

**Subject:** Re: Fw: FOR RELEASE MONDAY: GWMA release  
**To:** CN=Marianne Holsman/OU=R10/O=USEPA/C=US@EPA  
**Cc:** "Mike Bussell" <Bussell.Mike@epamail.epa.gov> CN=Caryn Sengupta/OU=R10/O=USEPA/C=US@EPA "Cox Michael" <Cox.Michael@epa.gov> CN=David Allnutt/OU=R10/O=USEPA/C=US@EPA CN=Jeff Philip/OU=R10/O=USEPA/C=US@EPA "Kate Kelly" <Kelly.Kate@epamail.epa.gov> CN=Marie Jennings/OU=R10/O=USEPA/C=US@EPA CN=Mark Macintyre/OU=R10/O=USEPA/C=US@EPA CN=Sheila Fleming/OU=R10/O=USEPA/C=US@EPA CN=Thomas Eaton/OU=R10/O=USEPA/C=US@EPA CN=Wenona Wilson/OU=R10/O=USEPA/C=US@EPA Cox.Michael@epa.gov  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 11/22/2011 22:50:29

Good questions.

### Results

- I do not think we want to provide a summary of results at this point as we are still going through internal EPA review and will be conducting a third party review in early December.

### Data to dairies, landowners, and homeowners

- Per advice from ORC, as soon as possible after laboratory validation we need to provide the data to the businesses or homeowners where we sampled. We are in the final process of doing this.

### FTP site

- In addition to providing data to the places we sampled, we are also posting all the data to an FTP site. We decided to post all the data to the FTP in order to be as transparent as possible.

### Messages

- We do need to work on the messages and planning for the final report and have started that process.

Hope this helps explain, along with bullets below, the status of the project.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Marianne Holsman---11/22/2011 01:35:12 PM---So, can someone summarize generally what these recent well results showed? I think we need to get to

From: Marianne Holsman/R10/USEPA/US  
To: Jeff Philip/R10/USEPA/US@EPA, Michael Cox/R10/USEPA/US@EPA  
Cc: Thomas Eaton/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA, Marie Jennings/R10/USEPA/US@EPA, Wenona Wilson/R10/USEPA/US@EPA, David Allnutt/R10/USEPA/US@EPA, Mark Macintyre/R10/USEPA/US@EPA, Caryn Sengupta/R10/USEPA/US@EPA, "Cox Michael" <Cox.Michael@epa.gov>, "Mike Bussell" <Bussell.Mike@epamail.epa.gov>, "Kate Kelly" <Kelly.Kate@epamail.epa.gov>  
Date: 11/22/2011 01:35 PM  
Subject: Re: Fw: FOR RELEASE MONDAY: GWMA release

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So, can someone summarize generally what these recent well results showed?

I think we need to get together and talk about communication planning going forward. It seems we're doing the messaging and planning on this one after the fact.

Marianne Holsman  
Via Blackberry  
Please excuse typos and possibly impolite brevity  
Jeff Philip

----- Original Message -----

**From:** Jeff Philip  
**Sent:** 11/22/2011 01:30 PM PST  
**To:** Michael Cox  
**Cc:** Thomas Eaton/R10/USEPA/US@EPA.; Sheila Fleming; Marie Jennings; Wenona Wilson; David Allnutt; Marianne Holsman; Mark Macintyre; Caryn Sengupta; Cox.Michael@epa.gov  
**Subject:** Re: Fw: FOR RELEASE MONDAY: GWMA release  
Thanks, Mike. This looks really good. Thanks for putting it together.

-----  
Jeff Philip  
Unit Manager  
Community Involvement & Public Information Unit  
U.S. Environmental Protection Agency, Region 10  
Seattle, Washington  
206.553.1465  
206.388.7505 cell

<http://epa.gov/region10>

Please save natural resources by not printing this email  
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Michael Cox---11/22/2011 08:51:53 AM---Not sure if we need all this, but pulled together. This is just for internal use. I am sure we cou

From: Michael Cox/R10/USEPA/US  
To: Jeff Philip/R10/USEPA/US@EPA  
Cc: Thomas Eaton/R10/USEPA/US@EPA., Sheila Fleming/R10/USEPA/US@EPA, Marie Jennings/R10/USEPA/US@EPA, Wenona Wilson/R10/USEPA/US@EPA, David Allnutt/R10/USEPA/US@EPA, Marianne Holsman/R10/USEPA/US@EPA, Mark Macintyre/R10/USEPA/US@EPA, Caryn Sengupta/R10/USEPA/US@EPA, Cox.Michael@epa.gov  
Date: 11/22/2011 08:51 AM  
Subject: Re: Fw: FOR RELEASE MONDAY: GWMA release

Not sure if we need all this, but pulled together. This is just for internal use. I am sure we could edit, but not sure if necessary at this point. I think Tom will be able to address any questions.

**Lower Yakima Valley Nitrates (November 22, 2011)**

**Talking Points**

- EPA provided results of sampling to all the participants in the study (dairies, landowners, wastewater treatment plants, and 24 residential drinking water users).
- Results are posted on FTP site.
- Final report will be completed by early 2012.
- EPA's role in GWMA is to support State and County.
- If questions contact either Caryn Sengupta or Tom Eaton.

### What was the study?

EPA conducted sampling in the Lower Yakima Valley between February – April 2010. The purpose of the sampling was to identify sources of nitrate contamination in groundwater in the Lower Yakima Valley. EPA analyzed for about 190 chemicals, plus nitrate, *E. coli*, fecal coliform bacteria, and total coliform bacteria. Many of the 190 chemicals analyzed are found where nitrate is found and may travel in groundwater in similar ways. EPA sampled at multiple locations including: dairies, irrigated croplands, domestic drinking water wells, and wastewater treatment plants.

What are the results, where can I access the results, and when will a report be completed?

EPA is still evaluating the results and anticipates a final report to be completed by early 2012. EPA has provided the results to all the locations where we collected samples. The letters to residents indicate whether or not any chemical found in their well exceeds the drinking water standard for that chemical. If the drinking water standard, also known as the Maximum Contaminant Level, was exceeded the letter provides health effects information for that chemical and what the resident can do to treat their water. Many of the chemicals analyzed for do not have drinking water standards or MCLs.

All of the data can be download from: [ftp://ftp.epa.gov/reg10ftp/sites/yakima/groundwater\\_data/](ftp://ftp.epa.gov/reg10ftp/sites/yakima/groundwater_data/).

Why is it taking so long to finish the report?

validated results in June 2011. The results from the laboratories took longer than anticipated for several reasons: 1) the analyses were complicated; 2) several of the laboratories had a high demand for their services and consequently our samples were delayed in being analyzed; and 3) problems with QA/QC at one lab required several months of back and forth and reanalysis of the pharmaceutical data.

**report prior to its release?**      **Who will be reviewing the**

The report is currently being reviewed internally within EPA. In early December, 4 third party reviewers will be provided with the draft report for review. EPA will incorporate the comments from the third party reviewers, complete the report, and then share with the public, government agencies, and agricultural/dairy community.

**contact if they have questions or concerns?**      **Who should people**

Tom Eaton has indicated he can respond to any questions about the release of the data and the GWMA. For general information people should contact Caryn Sengupta at 206-553-1275 or 1-800-424-4372 ext 0251. Julius Nwosu, toxicologist in OEA, is available to respond to questions from people concerned about the contaminants identified in their drinking water (206-553-7121).

**County lead GWMA?**      **What is EPA's role in the**

EPA's role is to participate on the working group that the State and County will be convening.

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US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Jeff Philip---11/21/2011 01:28:40 PM---Marianne brings up a good point. We should have talking points in the event a reporter calls regard

From: Jeff Philip/R10/USEPA/US  
To: Thomas Eaton/R10/USEPA/US@EPA., Sheila Fleming/R10/USEPA/US@EPA, Marie Jennings/R10/USEPA/US@EPA, Wenona Wilson/R10/USEPA/US@EPA, David Allnutt/R10/USEPA/US@EPA  
Cc: "Michael Cox" <Cox.Michael@epamail.epa.gov>, Marianne Holsman/R10/USEPA/US@EPA, Mark Macintyre/R10/USEPA/US@EPA  
Date: 11/21/2011 01:28 PM  
Subject: Re: Fw: FOR RELEASE MONDAY: GWMA release

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Marianne brings up a good point. We should have talking points in the event a reporter calls regarding the letters we sent out and their relationship to the GWMA.

Tom, would you be our spokesperson?

Can someone put together talking points on the results shared in the letters to residents this week?

We probably want to hit on a few issues:

What were the results shared in the letters?

Do our results indicate a larger problem with nitrates in groundwater?

Are there steps people should take based on the results?

What are EPA's next steps? (Final Report to be issued soon?)

What is EPA's role in the GWMA? Is there federal funding to support the GWMA?

---

Jeff Philip  
Unit Manager  
Community Involvement & Public Information Unit  
U.S. Environmental Protection Agency, Region 10  
Seattle, Washington  
206.553.1465  
206.388.7505 cell

<http://epa.gov/region10>

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Marianne Holsman---11/21/2011 12:40:33 PM---Hey Folks: Michelle P. asked me today about the sample results letters that were sent out and wanted

From: Marianne Holsman/R10/USEPA/US  
To: Mark Macintyre/R10/USEPA/US@EPA  
Cc: "Michael Cox" <Cox.Michael@epamail.epa.gov>, "Thomas Eaton" <Eaton.Thomas@epamail.epa.gov>, "Sandra Halstead" <Halstead.Sandra@epamail.epa.gov>, "Mr. Jeff Philip" <Philip.Jeff@epamail.epa.gov>, "Caryn Sengupta" <Sengupta.Caryn@epamail.epa.gov>  
Date: 11/21/2011 12:40 PM  
Subject: Re: Fw: FOR RELEASE MONDAY: GWMA release

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Hey Folks:

Michelle P. asked me today about the sample results letters that were sent out and wanted to make sure we had a desk statement/talking points ready in case we get any calls as a result. Do we have that? What did the results show?

Thank you.

\*\*\*\*\*

Marianne Holsman  
Public Affairs Director  
US EPA Region 10  
1200 6th Avenue  
Seattle, WA 98101  
desk: 206.553.1237 cell: 206.450.5895  
Email: holsman.marianne@epa.gov

Mark Macintyre---11/21/2011 11:35:07 AM----- Original Message ----- From: "Redfield-Wilder, Joye (ECY)" [JRED461@ECY.WA.GOV]

From: Mark Macintyre/R10/USEPA/US  
To: "Mr. Jeff Philip" <Philip.Jeff@epamail.epa.gov>, "Caryn Sengupta" <Sengupta.Caryn@epamail.epa.gov>, "Thomas Eaton" <Eaton.Thomas@epamail.epa.gov>, "Marianne Holsman" <Holsman.Marianne@epamail.epa.gov>, "Michael Cox" <Cox.Michael@epamail.epa.gov>, "Sandra Halstead" <Halstead.Sandra@epamail.epa.gov>  
Date: 11/21/2011 11:35 AM  
Subject: Fw: FOR RELEASE MONDAY: GWMA release

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**From:** "Redfield-Wilder, Joye (ECY)" [JRED461@ECY.WA.GOV]  
**Sent:** 11/21/2011 06:43 PM GMT  
**To:** Mark Macintyre  
**Subject:** FW: FOR RELEASE MONDAY: GWMA release

Mark – this News Release announcing formation of Yakima Nitrates GWMA will be out shortly. Joye

**From:** Redfield-Wilder, Joye (ECY)  
**Sent:** Monday, November 21, 2011 10:41 AM  
**To:** Partridge, Sandra (ECY)  
**Cc:** Redfield-Wilder, Joye (ECY); McKinney, Charlie (ECY); Tebb, G. Thomas (ECY)  
**Subject:** FOR RELEASE MONDAY: GWMA release

Sandra – this is ready to go to Yakima and Benton County media. Thanks - Joye[attachment "GWMA order NR edits.docx" deleted by Marianne Holsman/R10/USEPA/US]



**Subject:** Yakima Report: Summary and action items from Tuesday October 18th meeting  
**To:** CN=Caryn Sengupta/OU=R10/O=USEPA/C=US@EPA CN=Chan Pongkhamsing/OU=R10/O=USEPA/C=US@EPA CN=Curt Black/OU=R10/O=USEPA/C=US@EPA CN=DonaldM Brown/OU=R10/O=USEPA/C=US@EPA CN=Eric Winiecki/OU=R10/O=USEPA/C=US@EPA CN=Gina Grepo-Grove/OU=R10/O=USEPA/C=US@EPA CN=James Lopez-Baird/OU=R10/O=USEPA/C=US@EPA CN=Jennifer MacDonald/OU=R10/O=USEPA/C=US@EPA CN=Karma Anderson/OU=R10/O=USEPA/C=US@EPA CN=Marie Jennings/OU=R10/O=USEPA/C=US@EPA CN=Nicholas Peak/OU=R10/O=USEPA/C=US@EPA CN=Rochelle Labiosa/OU=R10/O=USEPA/C=US@EPA CN=Sandra Halstead/OU=R10/O=USEPA/C=US@EPA CN=Sheila Fleming/OU=R10/O=USEPA/C=US@EPA CN=Stephanie Bailey/OU=R10/O=USEPA/C=US@EPA CN=Stephanie Harris/OU=R10/O=USEPA/C=US@EPA CN=Steve Hutchins/OU=ADA/O=USEPA/C=US@EPA CN=Steven Potokar/OU=R10/O=USEPA/C=US@EPA CN=Theogene Mbabaliye/OU=R10/O=USEPA/C=US@EPA CN=Thomas Eaton/OU=R10/O=USEPA/C=US@EPA Cox.Michael@epa.gov  
**Cc:**  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 10/19/2011 17:44:34

Thanks for all those who attended and provided input. Unfortunately we lost the room at 3:00 so did not finish the discussion.

**Highlights:** I am sure there are many other comments that will come out that we will try to address.

1. **Comments due by noon Friday October 21:** Please give me whatever you have by noon on this Friday. I will be at a conference all next week and will be working on it during that time.
  2. Need to describe up front the genesis of the project (RARE) and the limitations this might impose (e.g., research methods). Also, need to be more specific on the lines-of-evidence that are potentially the most useful.
  3. Need to better explain the isotopic work including better explaining the relationship between perchlorate and isotopic analysis.
  4. Need to be more specific on which pharmaceuticals and hormones are used by humans and/or other animals. This will help better distinguish which sources main be contributing to the problems.
  5. Need to be consistent when describing the pharmaceuticals. Sometimes use veterinary pharmaceuticals and sometimes use other pharmaceuticals.
  6. Should not rule out dairies as a possible fertilizer source on crop fields.
  7. Need to better describe the purpose of the age dating and whether lagoons could be contributing to the "old" ages in the downgradient wells.
  8. Need to include any information we have on the well construction, depth etc in the report.
- Eric/Chan:** If you have any information on the wells please forward so we can include in the report.  
Thanks.

**Additional discussion needed**

1. Isotopic analysis - OEA will set-up conference call with Megan Young of USGS to discuss
  2. Need to decide whether to have another round of internal review prior to third-party review. If we have another round of internal review prior then we probably would not complete the report by the end of 2011.
- My suggestion:** Third-party reviewers are all federal employees. I would suggest sending out and then have a parallel review internally.



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Any correction, modifications, etc please call, email, or stop by.

Thanks.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

**Subject:** Re: Fw: Yakima Groundwater Report: Third Party Reviewers  
**To:** CN=Roger Burke/OU=ATH/O=USEPA/C=US@EPA  
**Cc:**  
**From:** CN=Stephen Kraemer/OU=ATH/O=USEPA/C=US  
**Submit Time:** 12/21/2011 21:51:40

It looks interesting. Something I could sit down with Ex. 6 Personal Privacy (PP)

Roger Burke---12/21/2011 04:19:00 PM---Steve, Please feel free to go ahead and send the report materials to me. I will probably be able to

From: Roger Burke/ATH/USEPA/US  
To: Stephen Kraemer/ATH/USEPA/US@EPA  
Date: 12/21/2011 04:19 PM  
Subject: Re: Fw: Yakima Groundwater Report: Third Party Reviewers

---

Steve,

Please feel free to go ahead and send the report materials to me. I will probably be able to help with the review but would like to do a quick evaluation of it to make sure I don't bite off more than I can chew.

Roger

Roger Burke  
USEPA/NERL  
960 College Station Rd.  
Athens, GA 30605  
Tel. 706 355 8134  
FAX 706 355 8104  
<http://www.epa.gov/athens/staff/members/burkerogera/index.html>

Stephen Kraemer---12/21/2011 04:00:08 PM---Roger, EPA Region 10 is looking to get third party review on their report "Relation between Nitrates

From: Stephen Kraemer/ATH/USEPA/US  
To: Roger Burke/ATH/USEPA/US@EPA,  
Date: 12/21/2011 04:00 PM  
Subject: Fw: Yakima Groundwater Report: Third Party Reviewers

---

Roger,

EPA Region 10 is looking to get third party review on their report "**Relation between Nitrates in Water Wells and Potential Sources in the Lower Yakima Valley, Washington**". I told Candida I would do the review but also look to recruit other expertise once the report was in our possession. It is my understanding that the report was produced by ORD/GWERD-Ada, OK. The main report is 77 pages long, and is missing the executive summary and some appendices. They would like comments by **Jan 13**.

I am going Ex. 6 Personal Privacy (PP)  
and my week back is filled with hydraulic fracking meetings in DC.

I am wondering if you might be interested in considering helping me with this review (I could cover parts of the report)? There is interesting analytical chemistry and isotope work. There is also field reconnaissance and GIS work that interest me. If this is a non starter for you just say no. But if curious, I could forward you the report materials so you can make a final decision.

Steve

----- Forwarded by Stephen Kraemer/ATH/USEPA/US on 12/21/2011 03:50 PM -----

From: Michael Cox/R10/USEPA/US  
To: Stephen Kraemer/ATH/USEPA/US@EPA  
Cc: Roseanne Lorenzana/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA, Cox.Michael@epa.gov  
Date: 11/22/2011 04:52 PM  
Subject: Yakima Groundwater Report: Third Party Reviewers

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Stephen,

We are going through another round of internal EPA review of the report and anticipate that we will get you a copy for review on Wednesday December 7th. We are asking, if possible, to provide your comments back by Friday January 7th.

I understand the holidays are upon us, but hope the one month review period will allow you enough time to review.

Thanks. Any questions please email or call.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Michael Cox---10/12/2011 01:49:36 PM---Stephen, Just getting back to you on the third party review. We are still in the process of develop

From: Michael Cox/R10/USEPA/US  
To: Stephen Kraemer/ATH/USEPA/US@EPA  
Cc: Roseanne Lorenzana/R10/USEPA/US@EPA, Cox.Michael@epa.gov  
Date: 10/12/2011 01:49 PM  
Subject: Re: Fw: Yakima Groundwater: Third Party Reviewers

---

Stephen,

Just getting back to you on the third party review. We are still in the process of developing the draft report and hope to have it ready for review by the first week in November. I am hoping this timeframe will work for you.

Thanks.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Stephen Kraemer---09/02/2011 06:08:45 AM---Michael. I know Dr. Hutchins and Dr. Burden well. I started my career at the Kerr Lab (1990-1997).

From: Stephen Kraemer/ATH/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Roseanne Lorenzana/R10/USEPA/US@EPA, MikeO Cox/R10/USEPA/US@EPA  
Date: 09/02/2011 06:08 AM  
Subject: Re: Fw: Yakima Groundwater: Third Party Reviewers

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Michael.

I know Dr. Hutchins and Dr. Burden well. I started my career at the Kerr Lab (1990-1997). That will not effect my objectivity. Depending on the content of the report, I think we may take a team approach in the review out here.

Cheers,  
Steve

Michael Cox---09/01/2011 06:39:47 PM---Steve, Looks like we will not have a draft for review until probably the middle of October. I wi

From: Michael Cox/R10/USEPA/US  
To: Stephen Kraemer/ATH/USEPA/US@EPA  
Cc: Roseanne Lorenzana/R10/USEPA/US@EPA, MikeO Cox/R10/USEPA/US@EPA  
Date: 09/01/2011 06:39 PM  
Subject: Re: Fw: Yakima Groundwater: Third Party Reviewers

---

Steve,

Looks like we will not have a draft for review until probably the middle of October. I will get a draft table of contents to you next week.

Just an FYI, we have been working with Steve Hutchins in Ada on this project and working on another project in the same area with Dave Burden. As Roseanne indicated below, we were hoping to get someone to provide third party review who had not been involved.

Thanks for help. I think it will be interesting

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US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Stephen Kraemer---08/31/2011 01:26:58 PM---Mike, Candida has assigned me to be your primary contact.

From: Stephen Kraemer/ATH/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Roseanne Lorenzana/R10/USEPA/US@EPA  
Date: 08/31/2011 01:26 PM  
Subject: Re: Fw: Yakima Groundwater: Third Party Reviewers

---

Mike,

Candida has assigned me to be your primary contact.  
The sooner I can see an abstract/executive summary/table of contents, the better I can plan a review approach.

Cheers,  
Steve

---

Stephen R. Kraemer, Ph.D., Research Hydrologist  
US EPA, Office of Research and Development (ORD)  
National Exposure Research Laboratory (NERL)  
Ecosystems Research Division (ERD)  
960 College Station Road, Athens, GA 30605-2700  
voice: 706-355-8340 fax: 706-355-8302

Candida West---08/31/2011 02:59:00 PM---Steve, Yes, this is the contact. You can go ahead and call Mike Cox to get details. Apparently NRM

From: Candida West/ATH/USEPA/US  
To: Stephen Kraemer/ATH/USEPA/US@EPA  
Cc: JohnM Johnston/ATH/USEPA/US@EPA, Roy Sidle/ATH/USEPA/US@EPA  
Date: 08/31/2011 02:59 PM  
Subject: Re: Fw: Yakima Groundwater: Third Party Reviewers

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Steve,  
Yes, this is the contact. You can go ahead and call Mike Cox to get details. Apparently NRMRL has been heavily involved and now they want to get an "outside" perspective. The work is envisioned to be two days of review, one day writing over about three weeks. Let us know how this develops.  
Thanks,  
Candida

---

Candida West, Ph.D.  
Deputy Division Director  
Ecosystems Research Division of the  
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U.S. Environmental Protection Agency  
Athens, Georgia 30605  
(706) 355-8023  
(706) 355-8007 (fax)  
West.Candida@epa.gov

Stephen Kraemer---08/31/2011 02:13:24 PM---I believe this is the call that you were talking about.  
Roseanne left a message for me, and then I

From: Stephen Kraemer/ATH/USEPA/US  
To: JohnM Johnston/ATH/USEPA/US@EPA  
Cc: Candida West/ATH/USEPA/US@EPA  
Date: 08/31/2011 02:13 PM  
Subject: Fw: Yakima Groundwater: Third Party Reviewers

---

I believe this is the call that you were talking about. Roseanne left a message for me, and then I left a message for her. Good she talked to Candida. I bet a team of ERD folks could provide the review. The other person might be Mohamed Hantush of NRMRL.

----- Forwarded by Stephen Kraemer/ATH/USEPA/US on 08/31/2011 02:11 PM -----

From: Roseanne Lorenzana/R10/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Roy Sidle/ATH/USEPA/US@EPA, Candida West/ATH/USEPA/US@EPA, Stephen Kraemer/ATH/USEPA/US@EPA  
Date: 08/30/2011 04:55 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

---

Mike:

In case you didn't get my voicemail, the ORD National Exposure Research Lab contacts:

Roy Sidle, Division Director 706-355-8001  
Candida West 706-355-8023  
Steve Kraemer 706-355-8340

These folks will help pin down exactly who can help with the review.  
Roseanne

-----  
Roseanne M. Lorenzana, DVM, PhD  
Diplomate of the American Board of Toxicology  
USEPA Region 10 Science Liaison  
1200 Sixth Avenue, OEA-095, Suite 900  
Seattle, Washington USA 98108  
206-553-8002 phone  
206-553-0119 fax  
lorenzana.roseanne@epa.gov  
<http://www.epa.gov/region10/>  
<http://www.epa.gov/ord/npd/>  
<http://intranet.epa.gov/ospintra/scienceportal/htm/regionalscience.htm>

Michael Cox---08/23/2011 02:30:31 PM---Yeah I would say someone should be able to review in two-days. Michael Cox Office of Environmental A

From: Michael Cox/R10/USEPA/US  
To: Roseanne Lorenzana/R10/USEPA/US@EPA  
Cc: Curt Black/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
Date: 08/23/2011 02:30 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

---

Yeah I would say someone should be able to review in two-days.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Roseanne Lorenzana---08/23/2011 02:26:33 PM---Hi Mike - I have been checking around. Discussion typically comes to the question of how many work

From: Roseanne Lorenzana/R10/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Curt Black/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
Date: 08/23/2011 02:26 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

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Hi Mike - I have been checking around. Discussion typically comes to the question of how many work hours are involved. Are you able to estimate? Would 16-24 hours be a reasonable estimate?

Roseanne

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*Roseanne M. Lorenzana, DVM, PhD, DABT  
Science Liaison to the Office of Research and Development  
U.S. EPA Region 10  
1200 Sixth Avenue, Suite 900, mail stop OEA-095  
Seattle WA 98101  
206-553-8002 phone 206-553-0119 fax  
lorenzana.roseanne@epa.gov  
<http://www.epa.gov/region10/>  
<http://www.epa.gov/ord/npd/>  
<http://intranet.epa.gov/ospintra/scienceportal/htm/regionalscience.htm>*

---

Michael Cox---08/23/2011 02:16:29 PM---Roseanne,

**Ex. 6 Personal Privacy (PP)**

**Ex. 6 Personal Privacy (PP)**

From: Michael Cox/R10/USEPA/US  
To: Roseanne Lorenzana/R10/USEPA/US@EPA  
Cc: Sheila Fleming/R10/USEPA/US@EPA, Curt Black/R10/USEPA/US@EPA  
Date: 08/23/2011 02:16 PM  
Subject: Yakima Groundwater: Third Party Reviewers

---

Roseanne,

I was checking if you had a chance to see if anyone from ORD maybe interested in participating in a third party review for the Yakima Report?

Thanks.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900

Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov





**Subject:** Re: Yakima Report: Draft Final for Review - Due date COB Thursday March 1st  
**To:** CN=Steve Hutchins/OU=ADA/O=USEPA/C=US  
**Cc:**  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 2/21/2012 20:43:44

I will send you the third party review comments tomorrow when I am back in the office.

Michael Cox  
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Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Steve Hutchins---02/18/2012 11:13:17 AM---Mike - I may have missed something - has this report already been sent out for peer review, and are

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\*\*\*\*\*

Stephen R. Hutchins, Ph.D.  
Research Environmental Scientist  
USEPA National Risk Management Research Laboratory  
Ground Water and Ecosystems Restoration Division  
Robert S. Kerr Environmental Research Center  
919 Kerr Research Drive, Ada, OK 74820  
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email: hutchins.steve@epa.gov

Michael Cox---02/16/2012 07:47:34 PM---Enforcement Confidential - Do Not Quote or Cite - Deliberative Process I am attaching several docume

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**James:** Figures

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**3. Appendix C - Sampling Results**

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## **5. Timetable**

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**Cc:**  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 2/22/2012 14:20:30

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Report December 16th 2011\_LE



Review\_Tarkalson.docx Lower Yakima Valley Draft

edits.docx  
Report December 16th 2011\_LE



Lower Yakima Valley Draft

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ke\_review\_13JAN2012.doc McMahon et al 2008 WRR source nitrate public wells.pdf



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portReview\_12Jan12.doc



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I think you have the comments from Ken Forshay.

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MAT]

**UNITED STATES ENVIRONMENTAL PROTECTION AGENCY**  
NATIONAL EXPOSURE RESEARCH LABORATORY  
ECOSYSTEMS RESEARCH DIVISION  
960 COLLEGE STATION ROAD | ATHENS, GA 30605-2700

OFFICE OF  
RESEARCH AND DEVELOPMENT

13 January 2012

**MEMORANDUM**

**Subject:** Review of EPA report “Relation Between Nitrates in Water Wells and Potential Sources in the Lower Yakima Valley, Washington”, draft, December 16, 2011

**From:** Stephen Kraemer, Ph.D., Research Hydrologist  
Roger Burke, Ph.D., Research Chemist

**To:** Michael Cox  
Office of Environmental Assessment, USEPA Region 10

Thank you for the opportunity to review the subject draft report on the investigation on nitrate sources to drinking water wells in the Yakima Valley. The scope and detail of the report are impressive. The writing is very good. The figures and tables adequately constructed. We will address the charge questions to the external reviewers below.

**1. Are the purpose, scope, and objectives of the project clear?**

Yes. The purpose of the study is to collect data to investigate the contribution of various sources from nearby land uses to the observed high nitrate in drinking water wells. The objective was to sample and analyze likely sources of nitrate (dairies, irrigated croplands, and septic systems) and private residential drinking water wells for a variety of chemicals to determine if chemicals other than nitrate can be used to link the nitrate contamination in groundwater to specific sources. The scope of the study included an area approximately 40 miles long and 10-25 miles wide.

**2. Is it clear why we selected certain chemical classes (e.g., hormones) or analytical techniques (e.g., isotopic analysis) to serve as potential tracers for nitrate contamination?**

Yes. The report does a good job of presenting background information.

**3. Is the experimental design clear?**

Yes. Although see responses (4) and (6).

**4. Is the approach taken for evaluating the isotopic data reasonable given the results from the study and the literature on isotopic analysis (e.g.,  $\delta^{15}\text{N}$ -NO<sub>3</sub> water well values greater than 8.4‰ characterized as dominated by animal waste;  $\delta^{15}\text{N}$ -NO<sub>3</sub> water well values less than 2.0‰ characterized as dominated by fertilizer; and  $\delta^{15}\text{N}$ -NO<sub>3</sub> water wells values between 2.0‰ and 8.4‰ being characterized as isotopically in determinant as to animal waste and/or fertilizer)?**



It is reasonable to attribute N isotope values greater than 8.4 parts-per-mille to animal waste as long as the nitrate concentrations are highly elevated - which they are in all cases reported in this document as far as we can tell. Based on soil N isotope values, and the fact that denitrification also causes isotopic fractionation that causes nitrate to become more <sup>15</sup>N-enriched, we suspect that soil cycling (e.g., Table 8, WW-02) could also produce nitrate with N isotope values greater than 8.4 parts-per-mille, albeit the nitrate concentration would probably be much less than observed for <sup>15</sup>N-enriched samples reported in the present study.

**5. Are the conclusions supported by the results?**

Yes. The conclusions stay within the evidence of the investigation.

**6. Are there results which could be more strongly used to link nitrate contamination to sources?**

Source water delineation for the drinking water wells based on computer modeling would further refine the concept of “upgradient” and “downgradient” used in the study, similar to the work of McMahon et al. (2008). The land uses and nitrate sources could be classified within the estimated zone contributing water to the pumping wells and compared to the chemical signatures. The USGS study used MODFLOW/MODPATH for individual well analysis. A regional analytic element model, e.g., WhAEM2000, would be better suited for regional scale, multi-well capture zone delineation.

McMahon, P. B., J. K. Bohlke, L. J. Kauffman, K. L. Kipp, M. K. Landon, C. A. Crandall, K. R. Burow, and C. J. Brown (2008), Source and transport controls on the movement of nitrate to public supply wells in selected principal aquifers of the United States, *Water Resour. Res.*, 44, W04401, doi:10.1029/2007WR006252.

**7. Are the uncertainties adequately addressed and clearly articulated?**

Yes. There is a whole section devoted to study limitations and uncertainties, including lack of well log information on the private wells, evidence of mixing of waters, the use of non standard research analytical methods, and limitations on information from dairy operations.

**Relation between Nitrates  
in Water Wells and  
Potential Sources in the  
Lower Yakima Valley,  
Washington**

**Draft Report:  
December 16, 2011**

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APPENDIX C: Quality Assurance and Quality Control

APPENDIX D: Information on R&M Haak Dairy

APPENDIX E: Information on Dairy Cluster (under development)

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## Acronyms/Abbreviations

AOAC – Association of Analytical Communities

CAFO – Concentrated Animal Feeding Operation

CARE – Community Action for a Renewed Environment

CFC – Chlorofluorocarbon

CHHP — Center for Hispanic Health Promotion

DEHP - bis-(2-ethylehxl)-phthalate

DOH — Department of Health

DQO – Data Quality Objectives

DG – Downgradient

EJ — Environmental Justice

EPA – U.S. Environmental Protection Agency

GIS – Geographic Information System

GPS — Global positioning system

“J” value – Compound was positively identified, but the associated numerical value is an estimate

“JN” value – There is evidence that the analyte is present. The associated numerical result is an estimate

LG – Dairy lagoon

LOE – Lines of Evidence

MCL – Maximum Contaminant Level

MDL – Method Detection Limit

MEL – EPA’s Manchester Environmental Laboratory

mg/L – Milligrams per liter

MST – Microbial Source Tracking

NCEC – Northwest Communities Education Center

ng/g – Nanogram per gram

ng/L – Nanograms per liter

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N<sub>2</sub> – Nitrogen Gas

<sup>14</sup>N- Nitrogen 14

<sup>15</sup>N – Nitrogen 15

NH<sub>4</sub><sup>+</sup> - Ammonium

NO<sub>2</sub><sup>-</sup> - Nitrite

NO<sub>3</sub><sup>-</sup> - Nitrate

ND – Not detected

NS – Not sampled

NWQL — National Water Quality Laboratory

ORD – Office of Research and Development

PCB — Polychlorinated biphenyls

ppm – Parts per million

QA/QC – Quality Assurance/Quality Control

QAPP – Quality Assurance Project Plan

QC – Quality Control

“R” value – The data are unusable for all purposes

RARE – Regionally Applied Research Effort

REDOX – Oxidation/Reduction Potential

RSKERC – Robert S. Kerr Environmental Research Center

SDWA – Safe Drinking Water Act

SF<sub>6</sub> – Sulfur hexafluoride

SMOW – Standard Mean Ocean Water

SOP – Standard Operating Procedure

TKN – Total Kjeldahl Nitrogen

“U” Value – The analyte was not detected at or above the reported result

µg/kg – Micrograms per kilogram

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µg/L – Microgram per liter

UG – Upgradient

“UJ” Value – The analyte was not detected at or above the reported estimated results. The  
associated numerical value is an estimate of the quantitation limit of the analyte in the sample

UNL – University of Nebraska Laboratory

USGS – U.S. Geological Survey

VIRE – Valley Institute for Research and Education

WW – Water Well

WWTP – Wastewater Treatment Plant

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## **EXECUTIVE SUMMARY**

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## I. INTRODUCTION

This report presents the results for sampling conducted from February 2010 to April 2010 by the U.S. Environmental Protection Agency (EPA) in the Lower Yakima Valley in Central Washington State. The primary purpose of this study was to investigate the contribution of various sources from nearby land uses to the high nitrate levels in drinking water wells. The study looked at three likely sources of nitrate: dairies; irrigated cropland; and residential septic systems.

EPA used standard investigation and analytical methods as well as several analytical methods applied primarily for research. The sampling was conducted as part of an EPA Regionally Applied Research Effort (RARE) grant (EPA 2009). Additional funding was provided by EPA Region 10's Office of Compliance and Enforcement and by the Yakima Valley Environmental Justice (EJ) Showcase Community pilot program.<sup>1</sup>

EPA's sampling effort in the Lower Yakima Valley was partially in response to concerns raised by several agencies and community members who participated in the EPA Community Action for a Renewed Environment (CARE) cooperative agreement with the Northwest Communities Education Center (NCEC) in Yakima County, Washington. The objective of the cooperative agreement was to assist the Yakima Valley community to establish its priorities for environmental health concerns. There were numerous meetings held over a 2-year period from 2007 to 2009. One of the outcomes from the cooperative agreement was that community members identified their top three environmental health priorities as groundwater contamination, asthma, and children's exposure to pesticides.

In October 2008, the *Yakima Herald Republic* ran a series of articles titled "Hidden Wells, Dirty Water" in which it examined a long history of groundwater contamination affecting public and private drinking water wells, primarily in the Lower Yakima Valley. The reporter sent a letter requesting that EPA invoke Section 1431 of the Safe Drinking Water Act (SDWA) to address the problem. Section 1431 authorizes EPA to take action when a contaminant is present or may enter a public water system or underground source of drinking water that may present an imminent and substantial endangerment to human health.

EPA facilitated formation of a work group consisting of representatives from state and local agencies, EPA, and the community. The work group released a report in February 2010, "Lower Yakima Valley Groundwater Quality: Preliminary Assessment and Recommendations." One of the recommendations identified in the report was to conduct an investigation to gather information to try to link high nitrate levels in drinking water wells with potential sources.

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<sup>1</sup> The purpose of the RARE program is to provide EPA Regional Offices with support for near-term applied research projects and enhance interactions and connections between Regional staff and EPA's Office of Research and Development. The EJ showcase projects focus on communities experiencing disproportionate impacts from an environmental health burdens.



The report documented that groundwater data collected in the Lower Yakima Valley from 1990 to 2008 indicated that as many as 12 percent of private wells had nitrate levels above the drinking water standard for nitrate (10 milligrams per liter [mg/L]) and about 20 percent of private wells demonstrated bacterial contamination (WADOE 2010). Nitrate is a naturally occurring form of nitrogen that can be found at concentrations between 0.5 mg/L and to 1.1 mg/L in shallow groundwater (Nolan and Hitt 2003). Nitrate concentrations higher than this range typically indicate that human activities have contributed nitrate to the groundwater.

## II. PURPOSE AND SCOPE

As discussed above, the primary purpose of this study was to collect data to investigate the contribution of various sources from nearby land uses to the high nitrate levels in drinking water wells. The objective of this investigation was to sample and analyze likely sources of nitrate (dairies, irrigated croplands, and residential septic systems) and private residential drinking water wells for a variety of chemicals to determine if chemicals other than nitrate can be used to link the nitrate contamination in groundwater to specific sources. The analyses included chemicals that are expected to be associated with one or more of the likely sources, such as pharmaceuticals (both veterinary and human medications), personal care products, steroids and hormones, pesticides and herbicides, as well as other indicators of water quality.

The investigation also used microbial analysis to determine whether the drinking water wells were contaminated with fecal contamination. If the water wells were found to have fecal contamination, then Microbial Source Tracking (MST) was performed to identify the source (i.e., human or ruminant) of the fecal contamination. In addition, EPA performed isotopic analysis for the water wells to determine the general source, or combination of sources, of nitrates in the water wells. Finally, an age dating analysis was completed for the water wells to determine the time since infiltration of water into the water wells.

**Figure 1** provides a conceptual site model for the project. The conceptual site model (in conjunction with **Figure 2 – Nitrogen Cycle**) provides a graphic description of how nitrate can reach groundwater and eventually drinking water wells. This study evaluated three likely sources of the nitrate contamination in drinking water wells (dairies; irrigated cropland, and residential septic systems). The main sources of nitrogen from the dairies include: dairy waste lagoons; manure piles; and manure applied to crops. For irrigated cropland, the main source is the synthetic fertilizers applied to the land to promote plant growth. For septic systems, it is the human waste that can migrate from the septic systems into adjacent drinking water wells.

As described in **Figure 2 (Nitrogen Cycle)**, nitrogen is applied to the land from different sources. The different forms of nitrogen migrate through the unsaturated silts, sands, and gravels and arrives at the water table via different preferential pathways. The nitrogen is converted to nitrate through different chemical and biological processes. The nitrate can then be transported to drinking water wells where humans consume the water or the nitrate can migrate to surface waters (in this case the Yakima River).

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The scope of this study includes an area approximately 40 miles long ranging between 10 and 25 miles wide where EPA had previously collected several hundred samples from residential wells to determine nitrate concentrations. EPA identified areas with some of the highest nitrate concentrations to conduct additional sampling to determine whether other chemicals are traveling with the nitrate from the sources to the groundwater and drinking water wells. This report includes the results for the sampling of 29 wells (25 residential wells and four dairy supply wells), 11 dairy lagoons (15 samples), 11 soil samples (five at dairy application fields and six at irrigated/fertilized crop fields), five dairy manure samples, and three wastewater treatment plant (WWTP) influent samples. The sampling was conducted in April 2010.

There were several constraints in the study that are important to note. Drinking water samples were collected from existing wells. No new wells or sampling points were installed for this study. Because of this, information on the well depths, screened intervals, and construction details of the wells was generally unknown. In order to analyze for certain chemicals, such as pharmaceuticals and hormones, EPA or equivalent standard methods have not been developed so methods used primarily for research purposes were ~~utilized~~<sup>used</sup>. Finally, there was limited information regarding the dairy operations. EPA requested information on specific aspects of the dairy operations to develop a better understanding of their day-to-day operations, however the dairies did not provide this information. This information would have contributed to a more complete understanding of the dairies' practices and their use of specific chemicals. These study constraints and their implications on interpreting the results of this study are discussed in Section X.

### III. BACKGROUND

Nitrate is an inorganic compound that is a naturally occurring form of nitrogen that can be found at concentrations between 0.5 mg/L and ~~to~~ 1.1 mg/L in ~~unimpacted~~ shallow groundwater (Nolan and Hitt 2003). Nitrate concentrations higher than this range typically indicate that human activities have contributed nitrate to the groundwater. Nitrate is highly soluble in water and mobile in soil, which make it relatively easy for nitrogen from a variety of point and non-point sources to ~~leach~~ through the soil and into the groundwater as nitrate.

Nitrate is an acute contaminant. EPA has established a Maximum Contaminant Level (MCL) for nitrate in drinking water of 10 mg/L under the SDWA. EPA regulates nitrate in public drinking water systems because nitrate concentrations greater than the MCL may cause a number of health problems. Exposure to excess nitrate can result in methemoglobinemia (blue-baby syndrome) in infants and susceptible individuals, which can lead to death in extreme cases (Ward 2005). Methemoglobinemia is caused by the reduction of nitrate to nitrite in the body. Nitrite binds to hemoglobin and lowers the body's ability to carry oxygen in the blood. Some studies have shown a positive association between long-term exposure to nitrate in drinking water and risk of cancer and certain reproductive outcomes, while other studies have shown no association (Ward 2005).

**Commented [LE1]:** Should read leach "from" the soil if you really mean that. If you just mean move through the soil, then don't use "leach."

Several water quality investigations for nitrate over the last 30 years in the Lower Yakima Valley, including the 2002 investigation by the Valley Institute for Research and Education (VIRE) were summarized in a February 2010 report entitled “Lower Yakima Valley Groundwater Quality: Preliminary Assessment and Recommendation Document” prepared by the Washington State Departments of Agriculture, Ecology and Health; Yakima County Public Works Department; and EPA (WADOE 2010). The report found nitrate levels above the EPA MCL of 10 mg/L in about 12 percent of private wells.

Nitrate contamination in groundwater is primarily a health risk for rural populations in the Lower Yakima Valley who rely on private wells for drinking water. Public water systems test regularly for nitrate and the data are reported to the Washington State Department of Health. Monitoring those systems that meet the definition of “public water systems” falls under state or federal drinking water regulations. EPA defines a public water system under SDWA Section 1401(4) as amended by the 1996 SDWA amendments as:

A public water system is a system for the provision to the public of water for human consumption through pipes or other constructed conveyances, if such has at least fifteen service connections or regularly serves at least twenty-five people.

The State of Washington has established requirements for systems serving between 3 and less than 15 connections and fewer than 25 people. These water systems are called Group B (Chapter 246-291 of the Washington Administrative Code), and the state Department of Health (DOH) and local health jurisdictions share responsibility for administering Group B requirements. The DOH does not regulate wells with just one or two connections that are residential systems, but some local jurisdictions regulate these systems. In 2009, the governor and state legislature set a new direction for regulating Group B systems by eliminating all state funding for this program.

Owners of drinking water wells that have fewer than three service connections (for example, a single, residential well) are not required to regularly sample their drinking water for contaminants. However, the EPA and the Washington State Departments of Ecology and Health recommend that rural residents test their well water regularly. If residents choose to sample and find contamination levels that exceed the MCL, they are not required to take action to address the situation.

#### IV. NITROGEN CYCLE

Nitrogen is present in many chemical forms in the environment including organic nitrogen, ammonium ( $\text{NH}_4^+$ ), nitrite ( $\text{NO}_2^-$ ), nitrate ( $\text{NO}_3^-$ ), and nitrogen gas. **Figure 2** shows the nitrogen cycle (adopted from Pidriwny 2006). The processes of the nitrogen cycle transform nitrogen from one chemical form to another. Important processes in the nitrogen cycle include nitrogen fixation, mineralization, nitrification, and denitrification. The mobility of nitrogen is highly dependent on its form and the matrix it moves through. In soils, nitrate is the most mobile form of nitrogen, with the exception of the gaseous form because negatively charged soil particles repel the negatively charged nitrate (Frans 2000).

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Nitrogen is an essential nutrient critical to plant growth in the formation and function of cellular tissue, proteins, and reproductive structures. Nitrogen can be supplied to plants through the organic decomposition of plants or animal waste products or by the application of synthetic fertilizers.

Nitrogen gas composes about 78 percent of the atmosphere. Atmospheric nitrogen must be processed, or fixed, to be used by plants. Some fixation is done by lightning strikes, but the majority of fixation occurs by bacteria. Additional small quantities of nitrate may wash out of the atmosphere from aerosol salt particles from the ocean or dusts from arid regions, or from fossil fuel combustion.

Mineralization occurs when the organic nitrogen in the soil is converted by bacteria into ammonium ( $\text{NH}_4^+$ ). The ammonium is then converted to nitrites and then nitrates by bacteria through nitrification. The nitrates can then be converted back into nitrogen gas ( $\text{N}_2$ ) by bacteria through denitrification. Denitrification occurs in low oxygen conditions in the soil. In the absence of denitrification, nitrates move with the groundwater until the groundwater is taken up by plants, discharged to surface water, or extracted from a well.

In human-influenced systems, there are significant increases in the amount of nitrogen released to the soil and frequently leached into groundwater from various land uses, including application of synthetic fertilizers or animal waste. While many fertilizers may be composed of nitrate, urea or ammonia are often used. The urea and ammonia is ultimately converted to nitrate by soil bacteria. Animal wastes are another source of nitrogen frequently applied to the land or they can be directly deposited by animals. Infiltrating rain or irrigation water can push excess nitrogen into groundwater from each of these sources, unless it is picked up by plants while still in the shallow subsurface. For additional information on the nitrogen cycle, see Stumm and Morgan, 1996.

**Commented [LE2]:** Just checking to make sure you mean "leach" here. OK if you do

## V. STUDY AREA

The Yakima Basin is a watershed of great diversity in climate, vegetation, and land use. More than 30 percent of the Yakima Basin is forested, 30 percent is sage-steppe rangeland, and 28 percent is in agricultural production (Vaccaro and others 2009). The Yakima River flows from its headwaters near the Cascade Mountains crest to its mouth as where it joins the Columbia River, 160 miles to the east. In the rain shadow of the Cascades, precipitation diminishes to less than 9 inches annually (Yakima County 2011), and irrigation plays a key role in the viability of agriculture. A series of high mountain reservoirs capture snowmelt, which is released through the Yakima River into a complex set of irrigation diversions and canals throughout the basin. Irrigation is supplied to fields during the March through October growing season in a variety of methods including flood, furrow, sprinkler, and drip systems.

This investigation focused on a portion of the Yakima Basin referred to as the Lower Yakima Valley (see Figure 3). This broad valley is bounded by basalt ridgelines to the north and south, the Cascade Mountains to the west, and encompasses two counties (Yakima and Benton) and the

**Commented [LE3]:** Is there a Study Area boundary, or is it everything shown on Figure 3?

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million-acre Confederated Tribes and Bands of the Yakama Nation Reservation. The study area includes portions of the Toppenish Basin (western area) and the Benton Basin (eastern area) along the Yakima River. Together, both areas cover approximately 368,600 acres within Yakima County. The Lower Yakima Valley has about 75,000 people, of which about 30,000 use private, unregulated residential wells (WADOE 2010).

In Yakima County, poverty afflicts greater than 20 percent of the population, and a little more than 30 percent of adults have less than a high school diploma. Approximately 41 percent of the population is Hispanic/Latino, which is more than four times the state average of nearly 10 percent. American Indians and Alaskan Natives make up a little more than 5 percent of the county's population, which is three times the state average of almost 2 percent. English is not the primary language (written or spoken) in many households in the Lower Yakima Valley (U.S. Census 2000). Economic viability depends on high-value agricultural production, irrigation, and a reliable supply of farm laborers. Yakima County leads the nation in production of milk per cow and is a top producer of apples, pears, sweet cherries, mint, and hops in the country (U.S. Census 2000).

#### A. Western Study Area - The Toppenish Basin

The ~~Much of the~~ Toppenish Basin is within the boundaries of the Yakama Nation. Land ownership in the major floodplain of the Toppenish Basin is a checkerboard of Indian trust, Indian fee, and deeded (privately held) parcels. Land use in this area is mixed, with open range and agriculture predominating. The basin is bordered on the north by the Ahtanum Ridge and on the south by the Toppenish Ridge.

**Commented [LE4]:** Isn't this accurate? Not all of the drainage basin is within the reservation as shown on Fig 3

#### B. Eastern Study Area – The Benton Basin

The Benton ~~Basin~~ includes the non-reservation lands along the river and ~~to the~~ on the southeast side of the valley. Approximately 60 percent of the valley population resides in this area, which includes the Yakima County communities of Sunnyside, Granger, Grandview, and Mabton.

**Commented [LE5]:** ? but some of the Benton Basin is included in the reservation along the river

The Benton Basin lies in the southeastern part of the Lower Yakima Valley. The western boundary of the basin abuts the eastern boundary of the Toppenish Basin. The southern boundary is bordered by the Horse Heaven Hills, and the northeastern boundary generally follows the northern flank of the Cold Creek Syncline.

#### C. Geology, Hydrogeology, and Geochemistry of the Study Area

The information presented below, unless otherwise noted, is summarized from the USGS publication "Hydrogeologic Framework of the Yakima River Basin Aquifer System, Washington" (Vaccaro and others 2009).

The Toppenish and Benton Basins consists of fine- and coarse-grained sediments overlying a sequence of three major basalt flows (see **Figure 4** and **Figure 5** for a general overview of the hydrogeology for the Toppenish and Benton Basins). The structural setting for the study area is created by bounding ridges such as the Rattlesnake Mountains, Ahtanum Ridge, Toppenish

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Ridge, and Horse Heaven Hills. The uppermost basalts of the Saddle Mountain Unit of the Columbia River Basalt Group are typically exposed in these upland ridges. This unit averages more than 500 feet thick. The underlying Wanapum unit averages 600 feet thick. These units are separated by the Mabton Interbed with an average thickness of 70 feet.

**Commented [LE6]:** These last two sentences are not needed if the basalts are not discussed again by name. Just say how thick they are.

The valley is filled with a variety of sediments that pinch out along the flanks of the ridges. These sediments ~~range from include~~ Touchet Beds, loess and thick alluvial sands and gravels, and significant thickness of Ellensburg Formation. The thickness of these sedimentary units decreases from an average of more than 500 feet in the Toppenish Basin to less than 200 feet in the lower Benton Basin.

**Commented [LE7]:** A "from" needs a corresponding "to"

Water is found in fractures and interbeds bounded by clinkers (a mass of vitrified material ejected from a volcano) and may be ~~first~~ found at significant depths in the upland ridges, such as Horse Heaven Hills, and especially in the basalts. The water table approaches the surface as the valley is approached from these ridges. Near the Yakima River, it may be less than 10 feet to water, especially during the irrigation season.

**Commented [LE8]:** Don't use unless you are discussing time or are clearly moving along a line from one part of the basin to another. This wasn't clear, so I deleted it.

There are two main aquifer types underlying the study area. They include a surficial unconfined to semi-confined alluvial aquifer and an extensive basalt aquifer of great thickness underlying the sedimentary deposits. The basalt aquifer is believed to be semi-isolated from the surficial aquifer and stream systems. Groundwater flow within the surficial aquifer generally follows topography, with natural recharge occurring within the headlands and on the sides of the valley and discharge occurring to the Yakima River. Flow within the uppermost portions of the deeper basaltic aquifer ~~can similarly follow also generally follows~~ this pattern.

However, since the basalts extend to great depths, those deeper basaltic layers may convey waters across local flow divides to more regionally significant discharge locations such as the Columbia River. This produces a major flow direction from northwest to southeast as water moves down the valley parallel to the course of the Yakima River. Other, more localized directions of flow — typically at shallower depths in the uppermost sediments — tend to flow toward the Yakima River with components of flow northeast to southwest on the north of the river and southwest to northeast on the south.

It is likely that even more localized and minor components of flow are significantly modified by irrigation practices upland from the Yakima River. These modifications of groundwater flow are not true-present in the deeper basalt aquifer in the Benton Basin, where ~~regional flow may be more regional beneath structures such as Rattlesnake Ridge and more generally flowing flows generally~~ toward the Columbia River. In this upland area, predominant groundwater flow is from the northwest toward the southeast. Locally, the flow direction may be modified by geologic structures and by drains, ditches, canals, and other hydrologic features.

**Commented [LE9]:** These two paragraphs say the same thing: regional flow toward Columbia vs localized flow affected by irrigation, etc. They could be combined.

Sediments such as those shed by the ridges at the margins of the study area and those sediments deposited in the valley bottom by the Yakima River have an internal structure that strongly controls groundwater movement. As the water moves through these sediments, it tends to follow preferential flow paths composed of coarser sediments. Very frequently, there are 10- to 100-fold

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differences in velocities ~~from one area to another among aquifer materials of such contrasting grain sizes~~ (Freeze and Cherry 1979). These different preferential flow paths can have different water chemistry depending on their location below a source of contamination. A well that is located along a preferential flow path may draw a substantial portion of its water from a particular source. A well located on an adjacent, but different, preferential flow path may have markedly different chemistry. For this reason, it is anticipated that upgradient sources of nitrates could produce different downgradient effects (such as nitrate in water wells), even in wells from neighboring homes.

**Commented [LE10]:** Freeze and Cherry citation is about the materials, not this specific area

In addition to the variability caused by the physical ~~makeup characteristics~~ of the aquifer, many compounds react with the aquifer materials in a way that changes their mobility. Some compounds like nitrate or ions like chloride interact very little and are transported nearly as fast as the water itself flows in the aquifer. Chloride tends to minimally adsorb to the aquifer material. Nitrate similarly minimally adsorbs and does not break down unless it encounters areas with very low oxygen in the aquifer and high concentrations of denitrifying bacteria. Other compounds, such as iron or manganese, often participate in chemical reactions and can create immobile minerals, which can change their concentrations as measured in water wells in unpredictable ways (Fetter 1980).

**Commented [LE11]:** Refer back to Figure 1, which illustrates this point very explicitly

Organic compounds, which are any gaseous, liquid, or solid chemical compounds containing carbon, are typically less mobile in water than inorganic compounds. Organic compounds tend to adsorb to organic carbon in the aquifer material and may be degraded by bacteria and either disappear entirely or may be greatly reduced in concentrations. Even if not broken down, most organic compounds will move much slower than nitrate because they tend to adsorb to other organic matter in the aquifer. As a result, in general, they are unlikely to be transported as far or as fast as the nitrate (Stumm and Morgan 1996).

## VI. THREE STUDY PHASES

Sampling efforts conducted to date in the Lower Yakima Valley by various agencies and groups have focused on nitrate. While these studies have been useful to document the problem of high nitrate levels in groundwater and private wells, they did not evaluate the link between the various sources and the high nitrate levels. The objective of this study was to sample and analyze sources of nitrate (dairies, irrigated crop lands, and residential septic systems) and private residential drinking water wells for a variety of chemicals to determine if chemicals other than nitrate can be used to connect the nitrate contamination in groundwater and drinking water wells to specific sources. Also, the study used several other analytical techniques (i.e., microbial source tracking, isotopic analysis, and age dating) to evaluate the contribution of various sources to high nitrate levels in drinking water wells.

To accomplish these objectives, EPA designed a three-phased study within two contiguous segments of the Yakima River Basin extending approximately 40 miles from the town of Union Gap to the Yakima County line near the town of Byron. The upper segment comprises the entire

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Toppenish Basin, and the lower segment comprises the northern portion of the Benton Basin. The width of the study area was defined by the width of the Toppenish and Benton Basins along the selected segment, which varies between approximately 10 to and 25 miles (Figure 3).

The purpose of Phase 1 was to identify and map major sources of nitrate in the study area, based on historical records. In Phase 2, the residential wells in closest proximity to the potential sources were identified, sampled, and analyzed for nitrates using screening-level analytical protocols and confirmatory laboratory analysis.

**Commented [LE12]:** Because the purpose was not to quantify these sources, just to make sure you captured the major ones, I believe you need to de-emphasize the numbers in the following sections.

Phase 3 of this study involved using the results of Phases 1 and 2 to identify residential wells with high nitrate concentrations and locate upgradient nitrogen sources. Once these source areas were selected, Phase 3 involved the collection and analyses of numerous samples from the potential source areas, downgradient wells, and upgradient wells (which were not available in some areas). The following subsections provide details about each phase of the study.

#### A. Phase 1: Geographic Information System (GIS) Tool Development and Screening Analysis for Nitrogen Sources

The purpose of Phase 1 was to identify and map major sources of nitrate in the study area, based on historical records. Phase 1 included the development of a GIS tool to organize a large amount of historical information and allow the examination of the landscape for spatial patterns in that data. EPA used the GIS tool to identify sites to be sampled in Phases 2 and 3 of the project. The tool incorporates information from the Lower Yakima Valley about known nitrate, bacteria, and general chemistry data. It also includes information on locations of wells, parcels with septic systems, land elevation, depth to groundwater, crop type, estimated fertilizer application rates, dairy and animal feeding operation locations, roads, and an aerial photo layer.

Phase 1 included a screening analysis to determine the potential sources of nitrogen in Yakima County. The screening analysis, described in more detail below, combined information on land use with some simple calculations in order to estimate the amount of "potential nitrogen available" from several sources. The estimates indicate that three sources: dairies, irrigated cropland and septic wastewater, can account for as much as 98% of the nitrogen available to be delivered to the aquifer.

**Commented [LE13]:** Don't jump from this general statement to "63% ..." It implies a lot more precision and certainty than appropriate. This is where you need to say that the estimates are only for relative comparisons and to make sure you've captured all of the potentially significant sources.

The screening analysis showed that about 63 percent of the potential nitrogen available comes from dairies, about 32 percent from inorganic fertilizers applied to irrigated crops, about 3 percent from septic and wastewater systems, and the rest from relatively minor sources such as nitrogen deposited by precipitation (EPA 2011a). These estimates did not account for losses from various biological, physical, and chemical processes.

**Commented [LE14]:** Or something like this. It might actually help a lot to include a table showing these calculations in one place.

**Commented [LE15]:** Without the info I added above, at this point in the document, the reader has no idea what you mean by this term. You need to introduce the topic in general, then explain that you used some simple, but reasonable assumptions, and only then say what the resulting numbers were.

**Commented [LE16]:** This makes it sound like a much harder number than it really is

**Commented [LE17]:** This is one of the qualifying statements that needs to be read in advance of the numbers

Based on this screening, EPA focused the Phase 3 sampling on three sources: dairies; irrigated cropland; and residential septic systems. Although there are other sources of nitrogen in the Lower Yakima Valley, EPA focused on the three areas believed to have the largest potential nitrogen contribution (see Figure 6).

**Commented [LE18]:** Figure 6 needs to be way smaller and integrated into the text. It currently has a very low ratio of information per square inch!



EPA is working to further refine these estimates and evaluate nitrogen fate and transport, in a collaborative project between EPA and the U.S. Geological Survey (USGS). A report is due in the fall of 2012. The project focuses on better characterizing the sources of nitrogen applied to the land and the relationship between ~~increasing or decreasing nitroge~~changes in nitrogen loading on the land ~~and~~ to levels of nitrate in drinking water wells.

## 1. Dairies

~~A trend over the past several decades has led to larger and larger dairies~~

**Commented [LE19]:** Don't say a trend has led to larger dairies if what you mean is that the larger size IS the trend. Not sure if my paraphrasing below is correct, but I gave it a try

Average dairy size has increased significantly in the last several decades (EPA 1998). Currently, 68 dairies in the Lower Yakima Valley are registered with the Washington State Department of Agriculture (see Figure 8). These facilities have a total of approximately 133,000 milking animal units (EPA 2011a), an average of almost 2,000 milking animal units per dairy. Modern dairies generate large quantities of animal wastes, which must be managed appropriately to prevent pollution, including pollution of surface and groundwater. Greater concentrations of animals and the scarcity of available land have made it increasingly challenging to effectively manage animal wastes to prevent adverse impacts to public resources.

**Commented [LE20]:** Figure 6 was the last one mentioned. The order in the text needs to agree with the numbering system.

In addition to generating large quantities of manure, dairies also generate large amounts of liquid waste from cleaning activities. Liquid wastes are typically flushed into a series of lagoons before they are sprayed on ~~nearby~~ fields as fertilizer.

Dairy wastes contain key components of fertilizer, including nitrogen, phosphorous, and potassium. When used as a fertilizer, dairy wastes are often supplemented with synthetic fertilizer to meet specific nutrient needs of the crop being grown. In the lower parts of the Yakima Valley dairies are concentrated around the cities of Sunnyside, Grandview, Mabton and Granger, although some are in more ~~sparsely~~sparsely populated areas of the valley and on the Yakama Reservation.

The total annual nitrogen production associated with dairies in Yakima County, without accounting for estimated losses, is approximately 36 million pounds per year. ~~This amount was calculated by multiplying the number of dairy cows by the estimated nitrogen production rate per cow provided by the Washington Department of Agriculture (EPA 2011a).~~

**Commented [LE21]:** This is the type of info that needs to be included in the introduction explaining how these numbers were derived (before the pie chart.)

## 2. Irrigated Cropland

Yakima County is one of the world's most fertile growing regions, with more than 240,000 acres of cropland. Agriculture is the primary economic activity in Yakima County, accounting for approximately 70 to 80 percent of land use. Most of the crop land in the area is irrigated. The major irrigation districts include the Roza, Sunnyside Valley, Wapato Irrigation Project, Grandview, and Zillah. Major commodities grown in the valley include apples, alfalfa, corn for silage and grain, grapes, hops, cherries, and mint (see Figure 9).

**Commented [LE22]:** Figure 9 needs a great deal of simplifying and clarifying. There are just way too many categories and they are way too hard to tell apart. Get rid of those that are land uses and not crops (fallow, golf course, research.) Make sure to highlight the ones you used in the analysis (corn, hops, mint). Either lump the rest into some logical categories (flowers, nursery crops and opiates, assuming those are poppy fields, for example) OR keep them separate, but use a similar color family for them, and a different one for other groups. Right now all the pinks are too similar, all the baby blues are too similar, and so on. Nothing is emphasized and almost everything is hard to identify. Fix type in "wheet."

Inorganic fertilizers contain high amounts of nitrogen. Nitrogen application is essential to crop growth and development, but crops can be treated with more nitrogen fertilizer than they are able

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to take up through their roots. Application can result in excess nitrogen infiltrating through the soil below the root zone into the groundwater. However, it is not just the use of fertilizer that can contribute nitrate to the groundwater, but rather the amount, timing, frequency, and type of fertilizer, as well as the timing and amount of irrigation relative to the application of fertilizer. Other factors such as denitrification in the soil by microorganisms, soil type, and volatilization to the atmosphere, also affect the amount of nitrate in groundwater.

EPA estimates that about 18.5 million pounds of nitrogen are added to the land each year in the form of inorganic fertilizers in Yakima County. This estimate was derived by taking the total acreage for each crop in Yakima County in 2007 and multiplying the acreage by the Washington State University-recommended average nitrogen application rate for each crop (EPA 2011a). These rates are general and the specific application rates and management practices by farmers could vary greatly. In addition, this estimate is the amount of nitrogen that is potentially applied to the land and does not include the uptake by plants nor the nitrogen removed at harvest or returned to the soil as crop residue.

As with the estimates for the dairies, the estimates were used as a relative value to compare with other estimates to assist in the study design. EPA is working to refine these estimates in a separate work effort discussed above.

**Commented [LE23]:** This is really important. Needs to be stated at the very beginning of the discussion of the screening phase of the project

### 3. Septic Systems and Wastewater

Domestic wastewater is managed by city wastewater treatment plants in Yakima County, but a large percentage of the mostly rural population relies on septic systems (see Figure 10). As of 2009, there were about 22,000 septic systems registered with Yakima County (EPA 2011a). Septic systems in Yakima County are permitted based on the average number of occupants per home square footage. Septic system use could exceed the design capacity in poor rural areas. Poor or deferred maintenance also could lead to improperly functioning septic systems, which could lead to increased concentrations of nitrogen moving into groundwater.

There are 16 permitted wastewater treatment facilities in Yakima County (EPA 2011a).

~~Wastewater~~ As wastewater treatment facilities process and treat wastewater, ~~to they~~ produce biosolids, which are nutrient-rich organic material. After the solids have been processed and treated, they are recycled as fertilizer and soil amendment. There are about 200,000 pounds per year of biosolids applied in Yakima County, which includes biosolids imported from metropolitan municipalities in Western Washington State (EPA 2011a).

An estimated 1.4 million pounds per year of potential anthropogenic nitrogen was calculated by multiplying the 2007 population in Yakima County (234,564) by the rate of 6 pounds of nitrogen per person per year (EPA 2011a). This approach provides an overall estimate of 1.6 million pounds per year of nitrogen from biosolids and septic systems or about 3 percent from septic systems and wastewater.

#### 4. Other Sources

Other sources of nitrogen, which are considered relatively minor, include nitrogen deposited by precipitation and non-cropland application of fertilizer to lawns, public parks, and golf courses. Application of nitrogen fertilizers was not estimated for the dryland wheat crops grown in the valley since they are not irrigated and the natural precipitation for the area likely limits nitrate leaching potential.

**Commented [LE24]:** After the discussion of the separate pieces, this is where I would summarize the numbers and show the pie chart.

#### B. Phase 2: Identification of Wells with High Nitrate Concentrations

The objective of Phase 2 was to determine the extent and magnitude of nitrate contamination downgradient of likely sources and to provide residents with information on the nitrate levels in their drinking water wells. The GIS tool developed in Phase 1 was used to help identify sampling locations for Phase 2. The sampling took place between February 22 and March 6, 2010. This time period corresponds to the pre-irrigation season when the aquifer is least influenced by recharge with ~~irrigation~~-delivered surface water. **Figure 7** provides a map of the locations and nitrate concentrations for the Phase 2 sampling. **Appendix A16** contains a summary of the results for the compounds evaluated in Phase 2.

**Commented [LE25]:** This should be re-numbered since it follows discussion of Figs 8, 9

EPA developed a Quality Assurance Project Plan (QAPP) for Phase 2 (USEPA, 2010a). It identifies the data quality objectives, sampling process design, sample collection procedures, sample handling and custody requirements, analytical methods, instrument calibration, data management, and standard operating procedures for instrument calibration, shipping container preparation, and chain-of-custody process. The Center for Hispanic Health Promotion (CHHP), a local bilingual, bicultural organization affiliated with the Fred Hutchinson Cancer Research Center, was contracted to assist in recruiting residences for sampling, scheduling, and Spanish interpretation assistance.

A series of public meetings, newspaper articles, and radio announcements notified the community of the Phase 2 work. Samples were collected by two-person teams trained for the project. Sample teams verified access from the homeowner, collected a global positioning system (GPS) location at the well, and completed a data collection form developed by EPA. Each sampling team maintained a field logbook to document sampling activities. For each well, water quality parameters were measured in the field using a Horiba multi-parameter probe.

The parameters measured included dissolved oxygen, oxidation/reduction potential, total dissolved solids, pH, and temperature. Measurements were taken at 1-minute intervals. At 5 minutes, the sampling team used nitrate colorimetric test strips as a field screening tool to provide an indication of whether the water exceeded the MCL of 10 mg/L for nitrate. The Hach test strips measure nitrate concentrations in increments of 0, 1, 2, 5, 10, 20, and 50 mg/L. If the test strip indicated the water may exceed the MCL (10 mg/L), samples were collected for analysis by EPA's Manchester Environmental Laboratory (MEL).

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Samples submitted to the laboratory were also analyzed for enumeration and quantification of total coliform using EPA's mobile microbiology laboratory. If total coliform bacteria were present, the samples were also analyzed for *E. coli* and fecal coliform bacteria. If *E. coli* or fecal coliform ~~were~~ detected, an additional sample was collected from the same well and analyzed using Microbial Source Tracking. Microbial Source Tracking is a molecular technology tool capable of differentiating human and ruminant sources of fecal contamination.

**Commented [LE26]:** Does this refer only to cows in this context? Might be worth a footnote for the benefit of the non-biologists who don't remember what other animals this includes.

During the 2 weeks EPA was in the field, about 330 homes were visited and all were screened for nitrate levels using the Hach tests strip. Seventy of those homes, or just over 24 percent, were found to exceed the MCL of 10 mg/L for nitrate (**Figure 7**). The percentage of homes with nitrate levels above the MCL ~~in this study~~ may be higher than the 12 percent from historical records because the homes sampled in Phase 2 of this study were selected ~~because they are in close proximity to likely sources~~. This method of selection would bias the results compared with a study where the sampling locations were selected randomly. Another possible explanation is that the previous studies were completed several years ago and the area with nitrate levels above the MCL ~~could~~ ~~may~~ have increased ~~in size~~.

**Commented [LE27]:** Based on the test strips or on the follow-up lab sampling? Clarify since both were discussed above.

Eight wells, or 2 percent, were found to have fecal coliform bacterial contamination or contamination with *E. coli*. This result is less than the 20 percent frequency found in past studies. Residents were informed of the results from the test strips immediately. Residents of all of the homes with nitrate levels greater than 10 mg/L or with bacterial contamination were provided with written laboratory results.

The Phase 2 sampling was informative in several ways. The results confirmed that nitrate concentrations in many domestic drinking water wells were above the EPA drinking water standard of 10 mg/L and provided information to the residents on the levels of nitrate in their wells. In addition, the Phase 2 results were used to identify the Phase 3 sampling locations.

### C. Phase 3: Investigating Contribution of Sources to High Nitrate Concentrations in Drinking Water Wells

The objective of Phase 3 was to investigate the contribution of various sources from nearby land uses to high nitrate levels found in water wells using a wide array of sampling and analysis techniques. The wells shown in **Figure 7** with the highest nitrate concentrations were selected for more extensive Phase 3 sampling and analyses. In addition, the specific sources associated with each well were selected for Phase 3 sampling and analysis. Representative upgradient wells also were selected for the locations where they were available.

After selecting all the sampling points, EPA grouped them into three basic types of sources (dairy waste, septic system wastes, and irrigated/fertilized crops) at five general sampling areas shown in **Figure 11**. Each area contains a number of nitrate-contaminated residential wells (above MCLs) and potential sources. Two of these areas contain only dairy-farm contamination sources (such as manure piles, lagoons, and application fields), and the three other areas contain both septic systems sources and irrigated cropland sources. The three source types and five sampling areas are shown in the **Table 1** (see attachment). **Table 1** also illustrates how the experimental

**Commented [LE28]:** There are 3 distinct called-out areas shown in Figure 11. It would help to name all 5 and show them on the map.

This actually gets more confusing as I read along. 8 areas are described—2 dairy, 3 septic, 3 irrigated. Everything that is a "sampling area" on Table 1 should be labeled on the map.

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design of the study varied, depending on the waste source type (dairy, septic system, or irrigated cropland). In general:

1. Investigation of each of the two dairy waste areas included sampling one upgradient well and a number of downgradient wells that are associated generally, but not individually, to a number of waste samples collected from lagoons, waste application fields, and manure piles. The well and waste samples were analyzed for many different chemicals, microbiology and using several different analytical techniques. The data for the downgradient wells were compared to the data for the upgradient wells and the various waste sources to determine if the different compounds can be used to identify specific sources.

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2. The investigation of the three septic waste areas included sampling residential wells downgradient from septic systems. The chemicals detected in the downgradient wells were compared to samples collected from the influent to wastewater treatment plants (WWTPs) located in Toppenish, Mabton and Zillah. These WWTP influent samples were selected to be representative of the types of chemicals that could be released from residential septic systems.

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3. The investigation of the three irrigated cropland areas (hops, mint, and corn) included sampling a total of six downgradient wells. Each well was exclusively paired with a soil sample from a specific type of crop. The chemicals detected in the downgradient well were compared to the chemicals detected in the corresponding soil sample from each of the six cropfields.

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Each of the sampling areas was evaluated looking at five different groups of chemicals or analytical techniques. The five groups included: general chemistry, microbiology, and organic chemicals. In addition, the well samples, lagoons, and WWTP samples were evaluated using isotopic analysis and the water wells were evaluated using age dating techniques.

Commented [LE30]: Confusing. This is only 3, then two more are listed for some specific sample types. So a subset gets just the 3 types of analyses? Which subset? Or you could clarify that all samples (without being specific) get these 3 types and then the third sentence describes the ones that get extras.

### 1. Phase 3 Sampling Locations

EPA used the Phase 1 GIS tool, Phase 2 sampling results, and a set of selection criteria to identify 63 sampling locations for Phase 3 (see **Figure 11** for the location for each of the sampling sites). **Appendix A1** provides the sample location, sample location type, description of the sample medium, and a summary of analytes at each location.

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#### Criteria for Selection of Dairies and Associated Sampling Locations

EPA collected samples at seven dairies. Dairy selection was based on data from Phases 1 and 2 of the project using the following criteria:

- High concentration of animals per unit area of available land.

- Indication of over-application of nutrients to application fields based on information contained in Washington Department of Agriculture inspection reports (*need cite*)
- Relatively consistent direction of groundwater flow from season to season.
- Minimal upgradient nitrate sources.
- Existence of private drinking water wells along the downgradient side, or sides, of the dairy.
- History of nitrate levels above the MCL in downgradient drinking water wells.

**Commented [LE31]:** Explain. Are these specific fields associated with the dairies for land application of wastes? Are they used consistently or does the application rotate to different locations?

Samples were collected from dairy lagoons, manure piles, application fields, and supply wells associated with the dairies. Wells upgradient and downgradient of the dairies also were identified for sampling. One sample was collected at the influent to each lagoon, and two samples were collected at the outlet from the lagoons. The manure pile samples were collected on site at each dairy. The application field samples were collected at the nearest field where lagoon waste recently had been applied.

**Commented [LE32]:** Are these the ones labeled "soil" on the map? Should clarify that if so.

#### Criteria for Selection of Residential Septic System Areas and Associated Sampling Locations

Samples were collected from four private drinking water wells that had high nitrate concentrations in Phase 2 and are located downgradient of areas with a ~~concentration~~ high density of residential septic systems in close proximity to one another. Additionally, samples were collected from the influent stream to three small wastewater treatment plants in the Lower Yakima Valley (Zillah, Mabton, and Toppenish) to serve as a surrogate for septic system influent and to characterize compounds found in rural septic systems. The criteria used to select the water well sampling locations in the residential septic system areas included:

- High ~~concentration~~ density of homes not served by sanitary sewers.
- Relatively consistent direction of groundwater flow from season to season.
- Minimal upgradient nitrate sources other than septic.

**Commented [LE33]:** Nothing wrong with this, but "concentration" is already used a lot with regard to water quality.

#### Criteria for Selection of Irrigated Cropland Areas and Associated Sampling Locations

Soil samples were collected from two fields each of corn, hop, and mint. Corn, hop, and mint were the crops selected because they require significant nitrogen to produce the large amounts of plant biomass for yield in contrast with other crops such as tree fruit. Thirty sub-samples per field were collected and composited to obtain a representative soil sample. One well was selected for sampling for each crop that was downgradient to the field. The criteria used for the site selection for the three crop fields were as follows:

**Commented [LE34]:** This is what you should explain earlier

**Commented [LE35]:** Of X (weight units?)

- Presumed history of high fertilizer application rates and use of agricultural chemicals.
- Relatively consistent direction of groundwater flow from season to season.
- Minimal upgradient nitrate sources.
- History of nitrates levels above the MCL in downgradient drinking water wells.

## VII. PHASE 3: COMPOUNDS AND ANALYTICAL TECHNIQUES

EPA analyzed for nearly 200 chemicals and utilized ~~used~~ several analytical techniques to investigate the high levels of nitrate in water wells to likely nitrate sources. The chemical analyses and analytical techniques were grouped as follows: general chemistry; microbial data; organic compounds; isotopic analysis; and age dating/gas studies. Each of the five groups are evaluated independently in an effort to connect specific sources to the nitrate found in residential drinking water wells.

This section describes the analyses that make up each of the five groups, why each of the analyses was conducted, and the issues or challenges associated with specific analyses and techniques. The data for the study are summarized in appendices referenced in each section below. A discussion of the analytical results is included in Section IX.

### A. General Chemistry

The study evaluated four ~~areas~~ aspects of general chemistry: nitrate and other forms of nitrogen; major ions; minor and trace inorganic elements; and perchlorate. Each is discussed below.

#### 1. Nitrate and Other Forms of Nitrogen

Samples from all water wells were analyzed for nitrate. All water wells, dairy lagoons, and WWTP influent samples were analyzed by EPA's Manchester Laboratory for nitrate plus nitrite, ammonia, and total Kjeldahl nitrogen (TKN). TKN is the sum of organic nitrogen and free ammonia. EPA conducted this analysis to ensure all major forms of nitrogen were identified, regardless of what form the nitrogen took.

For example, nitrate is generally not detected in a dairy lagoon because the chemistry of the dairy lagoon is anoxic (lack of oxygen) based on the large amounts of organic matter present. Nitrogen is present in the dairy lagoon, but in the form of organic nitrogen and as ammonia or ammonium released by bacterial action on the organic matter. Some of this ammonia can volatilize from the surface of the dairy lagoon. The ammonia is then converted to nitrite and then nitrate once it comes into contact with oxygen and bacteria.

In addition, total nitrogen was measured for each sample. Total nitrogen is the sum of nitrate, nitrite, and TKN. The concentrations of total nitrogen can be compared and evaluated for

**Commented [LE36]:** Some missing words here. "To investigate links between the high levels ..." (??)

**Commented [LE37]:** What I would have found handy in this section = summary tables!  
1. List sample type (wells, lagoons, etc.) in column A followed by columns for each analysis type. Place X in boxes where appropriate.  
2. For each category (major ions, minor and trace organics, pesticides, etc) a table listing what those analytes are—as in Tables 2 and 3.

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patterns in concentrations between upgradient water wells and downgradient water wells with likely sources, such as dairy lagoons and manure stockpiles that receive dairy waste located between these points. The results for the water wells, dairy lagoons, and WWTPs are included in **Appendix A2**.

All manure piles and application field samples were analyzed by Cascade Analytical Laboratory for extractable nitrate, extractable ammonia, and total nitrogen by combustion. These analyses were conducted to provide an indication of fertilization practices and the potential for leaching of nitrogen from the fields containing manure or inorganic fertilizer and to quantify the potential for these materials to act as sources of nitrate to the groundwater. The nitrogen not taken up by the plants would be available for mobilization by infiltrating rainwater or irrigation and could be delivered to the groundwater. The results for the manure piles and application field samples are included in **Appendix A3**.

Nitrate was analyzed at three different labs for different purposes. Cascade Analytical Laboratory in Union Gap analyzed the water wells samples for nitrate using EPA Method 300.0. Method 300.0 provides for measurement of nitrate alone. Method 300.0 requires the sample to be analyzed within 48 hours after it is collected. This method was used for the water well samples because it is specified as the method for evaluating the MCL for nitrate and Cascade Analytical Laboratory was used because of its proximity to the study area. The short holding time for Method 300.0 (48 hours) made it a challenge to get the samples to EPA's Manchester Laboratory in the required time.

EPA's Manchester Laboratory analyzed for nitrate as part of the general chemistry suite using Method 353.3. Method 353.3 measures nitrate plus nitrite and is one of several analytes measured in Method 353.3. This method has a holding time of 28 days because the samples are preserved. Finally, the University of Nebraska analyzed samples for nitrate using Method 353.3 to complete the isotopic analysis.

**Appendix A4** provides a comparison of the nitrate concentrations reported by each of the three laboratories for the water wells sampled in Phase 3. The results for the nitrate analysis are similar among the three laboratories, which suggest the analyses are accurate. (One exception was sample WW-18.) Even for WW-18, two of the labs reported consistent concentrations of 72.2 mg/L (Cascade) and 72.3 mg/L (UNL).

#### **4.2. Major Ions**

All water wells, dairy lagoons, and WWTP influent samples were analyzed for the major ions by EPA's Manchester Environmental Laboratory. The major ions were not analyzed for soil and manure samples because, in general, the purpose for analyzing the major ions is to track the chemical evolution of migrating groundwater.

An ion is an electrically charged species consisting of a single atom or a group of atoms. It is formed when a neutral atom or group of atoms either gains or loses electrons. The major ions



evaluated included: calcium, chloride, fluoride, iron, magnesium, nitrate, potassium, sodium, and sulfate. The results for the major ions are included in **Appendix A5**.

Different ions have different chemistries and transport mechanisms. For example, chloride does not generally sorb to particles or participate in reactions with the aquifer material. Other ions, such as potassium and sodium, are much more likely to react with minerals and sorb to aquifer materials. Because of the absence of oxygen, sulfate would not be expected to be found in the dairy lagoons. The expected form of sulfur in the dairy lagoons would be sulfide or sulfur still bound in the organic matter. Sulfate would be formed after the dairy lagoon waste escapes from the lagoon and has the opportunity to react with oxygen, oxidizing the forms of sulfur to mobile sulfate in the groundwater.

For this study, the major ions were evaluated for spatial patterns in concentrations from upgradient wells to downgradient wells. If the concentrations in the downgradient wells are higher for specific ions than in the upgradient wells, and those same ions are abundant in a specific source such as a dairy lagoon, then the source is a likely contributor to those higher levels. For example, if chloride is detected at high levels in a dairy lagoon and the concentrations of chloride in a water well downgradient of the dairy lagoon are higher than in compared with a well upgradient of the dairy lagoon is higher, it is an indication the dairy lagoon is contributing chloride to the downgradient well.

### 5.3 Minor and Trace Inorganic Elements

All water wells, dairy lagoons, and WWTP influent samples were analyzed for minor and trace inorganic elements by EPA's Manchester Laboratory. Twelve minor and trace inorganic elements were evaluated: arsenic, barium, bromide, cadmium, chromium, copper, lead, manganese, mercury, selenium, silver, and zinc. Minor and trace inorganic elements were not analyzed in the soil or manure samples for similar reasons cited above for the major ions. The results for the minor and trace inorganic elements are included in **Appendix A5**.

The trace inorganic elements were included in this study because the sensitivity of certain metals to oxidation/reduction potential (how oxygen rich the waters are) can provide indications of releases from dairy lagoons. For example, the change in oxygen can lead to the increased mobility of metals such as arsenic and manganese. Oxygen is consumed in microbial reactions that use organic carbon. If an increased concentration of arsenic and manganese in downgradient water wells in comparison to upgradient water wells was seen, then it could provide an indication of the influence of an organic carbon source such as dairy lagoons.

**Commented [LE38]:** But it is not the "sensitivity" that gets measured. So I'm hoping the rest of the paragraph explains this.

**Commented [LE39]:** Specify increase or decrease since this is so specific

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### 5.4 Perchlorate

All wells were tested for perchlorate and analyzed by the EPA's Robert S. Kerr Environmental Research Center (RSKERC) in Ada, Oklahoma. The results for perchlorate are in **Appendix A6**. Perchlorate is the most highly oxidized form of chlorine and tends to accumulate in caliche associated soils in arid regions such as Eastern Washington and Oregon (Rao and others 2007). In this study, it was used as an indicator for potential naturally occurring nitrates.

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There is a very slight, but steady, deposition of nitrate and perchlorate from the atmosphere. Much of it starts as aerosol salt particles released from combustion in transportation or power generation or carried off the oceans as aerosols or dust particles from deserts by winds (Prospero and Lamb 2003). In this region, the National Atmospheric Deposition Program ([ HYPERLINK "http://nadp.sws.uiuc.edu/" ]) calculates aerial deposition of atmospherically derived nitrate at approximately 0.9 pounds per acre per year. Perchlorate accumulates at much lower rates but has not been studied to the same extent, so data are lacking.

This accumulation of nitrate and perchlorate has been occurring since the end of the last glacial period, approximately 10,000 years ago. In higher rainfall areas, both these compounds are sufficiently soluble to be carried into the subsurface and potentially into groundwater. However, these compounds can build up in the shallow subsurface with the calcium carbonate that forms the cement-like caliche layer in arid regions such as the Lower Yakima Valley.

The same conditions that would wash the nitrate out of a caliche soil horizon – the first application of irrigation water to a new field converted from sage habitat – would flush out perchlorate as well. These two compounds, nitrate and perchlorate, are somewhat differentially extractable and it is more common to find conditions where nitrate persists but the perchlorate has been mobilized and lost to the underlying groundwater. As shown in the results section, only a few of the wells showed high values of perchlorate despite the presence of significant amounts of nitrate with an isotopic signature suggesting an atmospheric origin.

**Commented [LE41]:** Are you trying to say the data are often difficult to interpret?

**Commented [LE42]:** Results are not discussed for other parameters in this section. Why for this one?

#### D.B. Microbiology

All water wells, dairy lagoons, and WWTP influent sample were analyzed for either total coliform, fecal coliform, or *Escherichia coli* (*E. coli*) as an indicator of fecal contamination. The results for microbiology are in **Appendix A7**. EPA's Manchester mobile microbiology laboratory or Cascade Analytical Laboratory in Union Gap conducted the analysis. Microbial Source Tracking (MST) was performed in samples that tested positive for fecal coliform. MST was not completed for the water wells because fecal coliform was not detected in any of the water wells. MST was not completed on nine of the 15 dairy lagoons ~~and either any of the~~ three WWTP influent samples because of limited resources.

MST is a means of identifying the source of the fecal contamination in a water sample. The method used in this study is genotypic and is used to detect the presence of host-specific *Bacteroides* species shed in the fecal material of humans or ruminants. This method allows a presence or absence reporting format for these two sources. A common way of referring to the host-specific genetic identifier for each of these species is a "biomarker."

Because this method is limited to presence or absence reporting for only human and ruminant sources, the data cannot be used to: (1) identify the quantity or proportional levels of contamination from either source; (2) identify specific sources other than human or ruminant; or (3) differentiate between the various kinds of ruminants — cattle, goats, sheep, deer, or elk.

**Commented [LE43]:** This is the explanation I was looking for the first time the word was used.

However, the data can be used to: (1) identify the frequency of identification of either of the sources from a particular sampling site if more than one set of samples is collected from the same site; (2) identify human or ruminant source contamination; and (3) confirm that recent fecal contamination has occurred.

## E.C. Organic Compounds

The study looked at four groups of organic compounds: pesticides; trace organics; pharmaceuticals; and hormones. Each of the four is discussed below.

### 1. Pesticides

Fifty pesticides were analyzed in water wells, dairy lagoons, WWTP influents, manure piles, and application field samples by EPA's Manchester Environmental Laboratory. The term "pesticide" refers to insecticides, herbicides, fungicides, and various other substances used to control pests. The pesticide analysis conducted as part of this investigation included insecticides and herbicides. The results for the pesticides are included in **Appendix A8**.

The pesticides selected for analysis were those that USGS reported had been used in the Yakima Valley and are considered mobile in groundwater, persistent, or both (Nakagaki and Wolock 2005). These compounds were considered possible tracers that might provide further information in support of the link of nitrate introduced with irrigated crop production and from field application of waste from dairies to the nitrate detected in water wells.

Many of the pesticides are used on specific crops and during specific times of the year. This pattern of usage can be an advantage as it can assist to identify the specific crop where the pesticide was applied. At the same time, it is possible that a particular pesticide, though used in the area, was not applied recently and so not detected in the soil samples collected by EPA because EPA collected samples over a limited time period (February to April).

EPA's Manchester Laboratory reported that the sample matrices provided significant interferences that made pesticide analysis difficult for dairy lagoons and WWTP influent samples. Because of this problem, the pesticide concentrations could not be quantified in the dairy lagoons or WWTP influent samples. The laboratory attempted to develop an extraction and cleanup procedure for the dairy lagoon and WWTP matrix; however, a procedure to resolve the matrix interference could not be developed within the maximum holding time specified for these samples. By the time the laboratory could have developed and tested an effective and reliable procedure, the maximum sample holding times would have been exceeded. Therefore, the pesticide results for the dairy lagoon and WWTP samples are considered unusable.

### Trace Organics

Each water well, dairy lagoon, and WWTP influent sample was tested for 67 trace organic compounds by the USGS National Water Quality Laboratory in Denver. The trace organics were

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not analyzed in soil or manure samples because the USGS laboratory was not equipped for this type of analysis and the methods for extraction of such samples are complex. The results for the trace organics and a description of their main use are included in **Appendix A9**.

The USGS developed a method for analyzing a large number of trace organics because they and other researchers had found them in domestic and industrial wastewater (Zaugg and others 2006) as well as groundwater and surface waters (Kolpin and others 2002; Barnes and others 2008). EPA believed the trace organics would help to differentiate water wells affected by septic systems (humans) from water wells influenced by other sources such as dairy lagoons or irrigated cropland. The compounds analyzed include many that can be associated with human usage, including caffeine, bisphenol A; cholesterol; menthol; phenol; various flame retardants; acetophenone (fragrance in detergent); benzophenone (fixative for perfumes); camphor (flavor, oxidant); isoborneol (fragrance in perfume); and many others.

### 8.3 Pharmaceuticals

The sample from each water well, dairy lagoon, WWTP influent, manure pile, and application field was analyzed for 14 wastewater pharmaceuticals (**Table 2**). The University of Nebraska Water Sciences Laboratory in Lincoln, Nebraska (UNL), performed the analysis. The results are included in **Appendix A10**.

**Table 2: UNL - Wastewater Pharmaceuticals Evaluated**

Compound Name	Description
Acetaminophen	Pain Reliever (Tylenol)
Amphetamine	Psychostimulant (Dexedrine)
Azithromycin	Antibiotics (Zithromax)
Caffeine	Stimulant
Carbamazepine	Anticonvulsant
Cotinine	Metabolite of nicotine
DEET	Insect repellent
Diphenhydramine	Antihistamine
Ibuprofen	Pain reliever
Methamphetamine	Psychostimulant
Naproxen	Pain reliever (Aleve)
Paraxanthine	Stimulant (metabolite of caffeine)
Thiabendazole	Parasiticide (mintezol)
Triclosan	Antibacterial

The group is identified as “wastewater pharmaceuticals” because they are generally used by humans for therapeutic reasons and have been detected in municipal wastewater (Temes and others 2004), surface waters (Kolpin and others 2002), groundwater (Barnes and others 2008), and drinking water (Benotti and others 2009). Many of the compounds are for over-the-counter

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use (for example, acetaminophen and ibuprofen) and are ingested, but a few are applied topically (DEET and triclosan). Two of the compounds can be used in other animals (thiabendazole and DEET).

Humans typically excrete 50 to 90 percent of the active ingredients in ingested drugs, either as unmetabolized pharmaceuticals or as metabolites (McGovern and McDonald 2003). These excreted compounds can enter a municipal WWTP or a septic system. Detection of these compounds in water wells may provide evidence that septic systems are the primary source of nitrate.

In addition, the sample from each water well, dairy lagoon, WWTP influent, manure pile, and application field was analyzed for 17 additional pharmaceuticals and ~~labeled-classified as~~ "veterinary pharmaceuticals" for this study. **Table 3** lists the compounds and the current FDA-approved uses (FDA, 2011(a) and FDA, 2011(b)). Many of the pharmaceuticals shown in **Table 3** do not require a veterinarian's prescription and are available for over-the-counter purchase (FDA, 2011(a) and FDA, 2011 (b)).<sup>2</sup> The majority of the over-the-counter pharmaceuticals are included in the feed for the animals. The University of Nebraska Water Sciences Laboratory in Lincoln, Nebraska, also conducted these analyses. The results are included in **Appendix A11**.

**Commented [LE45]:** but this is a good place to point out that several of the antibiotics are used by humans as well

Detections of the compounds in **Table 3** in water wells would provide evidence that dairies are a likely source of those compounds. For example, if monesin is detected in water wells, then it is coming from a source other than humans. (Monesin is not approved for use in humans.) If the compounds are detected in dairy lagoons, manure piles, or application fields, it is a good indication that the dairy is using the compound. If detected in the influent to the WWTPs, it can establish whether these compounds are being excreted by humans and ending up in municipal sewage waste. If the compounds are detected in the WWTP influent, they can be compared with detected compounds in water wells to evaluate whether septic systems may contribute to the presence of these compounds in well water.

**Table 3: Veterinary Pharmaceuticals Evaluated and FDA Approved Uses as of November 2011**

Compound Name	Current FDA Approved Use
Chlortetracycline (total)	Cattle (beef, dairy), poultry, swine, and sheep
Erythromycin	Cattle (beef, dairy) and humans
Lincomycin	Swine, poultry, and humans
Monesin	Cattle (beef, dairy), and poultry
Oxytetracycline	Cattle (beef, dairy), poultry, sheep, and humans
Ractopamine	Cattle (beef), swine, and poultry.
Sulfachloropyridazine	Cattle (beef), swine, and sheep

<sup>2</sup> Compounds that can be obtained over-the-counter include chlorotetracycline; erythromycin; lincomycin; monesin; ractopamine; sulfadimethoxine; sulfamethazine; sulfathiazole; tetracycline; tiamulin; trenbolone; tylosin; and virginiamycin.

Compound Name	Current FDA Approved Use
Sulfadimethoxine	Cattle (beef, dairy), and poultry
Sulfamerazine	Poultry
Sulfamethazine	Cattle (beef, dairy), poultry, and swine
Sulfamethizole	Dogs and cats
Sulfamethoxazole	Humans
Sulfathiazole	Swine
Tetracycline	Cattle (beef, dairy), poultry, sheep, swine, and humans
Tiamulin	Swine
Tylosin	Cattle (beef, dairy), poultry, and swine
Virginiamycin	Poultry, swine, and poultry

The UNL analyzed the compounds in **Table 3** because they are used in livestock production at therapeutic doses to treat and prevent disease and at sub-therapeutic doses as prophylactics and growth promoters (Meyer 2004) and have been found at low levels in different environmental media: groundwater (Barnes and others 2008 and Kummerer, 2009); surface water (Koplin and others 2002; Christian and others 2003; and Kummerer, 2009); and wastewater treatment facilities (Temes and others 2004; and Lubliner and others 2010). More specifically, several of the compounds have been found in dairy lagoons (Watanabe and others 2008 and Watanabe and others 2010); soil and surface samples from dairies (Watanabe and others 2010); private wells nearby a beef cattle operation (Batt and others 2006); and in groundwater underlying swine and beef cattle facilities (Bartlett-Hunt and others 2011). Some of the compounds in **Table 3** are used by humans (Kummerer 2009).

The U.S. Department of Agriculture's National Animal Health Monitoring System conducted a survey to evaluate the use of antibiotics in dairy operations for disease prevention, disease treatment, and growth promotion in preweaned heifers, weaned heifers, and mature cows (USDA 2008). The survey represented 17 of the nation's major dairy states (Washington was included) and represented about 82 percent of the U.S. dairy cows. The results indicate that the majority of dairy operations use antibiotics to treat for diarrhea, digestive problems, respiratory problems, mastitis, reproductive disorders, and lameness.

To identify which of the pharmaceuticals might be used by the dairies in this study, EPA requested information from the dairies on the use of pharmaceuticals in their operations. The dairies declined to provide this information to EPA and therefore there is no specific information on the use of these compounds by the dairies that are included in this project.

#### **9.4 Hormones**

Each water well, lagoon, and WWTP plant influent sample was analyzed for five hormonally active compounds (17-a-estradiol, 17-a-ethynyl-estradiol; 17-b-estradiol; Estriol; and Estrone) by EPA's Subsurface Characterization Laboratory in Ada, Oklahoma. The results for these hormones are in **Appendix A12**.

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In addition, each water well, lagoon, WWTP plant influent, and soil/manure sample was tested for 18 hormonally active compounds by the UNL Water Sciences Laboratory in Lincoln, Nebraska, including the five analyzed by Ada. The results for these analytes are in **Appendix A13**. **Table 4** shows all the compounds evaluated and their natural source or general use. The table also provides information on the FDA-approved uses for the analytes as of November 1, 2011 (FDA 2011a and FDA 2011b).

**Table 4: Ada and UNL – Hormonally Active Compounds Evaluated**

Compound Name	Description (Current FDA Approved Use)
<b>Analyzed at both Ada and UNL</b>	
17β-estradiol	Natural female sex hormone (beef cattle)
α-Estradiol	Predominant sex hormone in females (beef cattle)
Estril	One of three main estrogens produced in mammals
Estrone	One of three main estrogens produced in mammals
17-α-Ethynyl Estradiol	Synthetic analogue of estradiol (human)
<b>Analyzed at UNL Only</b>	
11-Keto Testosterone	Oxidized form of testosterone
17α-Hydroxyprogesterone	Natural progestogen
4-Androstenedione	Intermediate step in producing testosterone and estrogens
Androsterone	Metabolite of testosterone
Epitestosterone	Naturally occurring form of testosterone
Progesterone	Natural female sex hormone (beef cattle and human)
Testosterone	Natural male sex hormone (beef cattle and human)
17α-trenbolone	Synthetic growth promoter (beef cattle)
17β-trenbolone	Synthetic growth promoter (beef cattle)
Androstenedienedione	Precursor to boldenone (boldenone: horse)
α-Zearalanol	Metabolite of zeranol (zeranol: beef cattle and sheep)
α-Zearalenol	Metabolite of zeranol (zeranol: beef cattle and sheep)
β-Zearalanol	Metabolite of zeranol (zeranol: beef cattle and sheep)
β-Zearalenol	Metabolite of zeranol (zeranol: beef cattle and sheep)
Melengesterol Acetate	Synthetic growth promoter (beef cattle)

UNL developed a method for analysis of these hormones in order to detect the compounds at the low levels of detection in environmental samples. Many of the hormones are naturally excreted by animals and can be used as pharmaceuticals in human and veterinary clinical practices (Zheng and others 2007). Many of the compounds have been detected at low levels in different environmental media or sources including: surface waters (Kolpin and others 2002); dairy lagoons (Kolodziej and others 2004; Aron and others 2008; Hutchins and others 2007, and Zheng and others 2008); groundwater associated with dairies (Kolodziej and others 2004 and Aron and others 2008); and manure at dairy facilities (Raman and others 2004).

Since many of the compounds are produced naturally by both humans and animals, it is challenging to identify the source of the hormones if detected in water wells. One compound (17- $\alpha$ -ethynyl-estradiol) is a synthetic analogue of estradiol that is primarily used in hormonal contraception and would not be expected to be found in lagoons, unless the lagoons are impacted by human waste, but maybe found in WWTP influent and septic systems, given its use by humans.

Several of the compounds analyzed by UNL are synthetic growth hormones (e.g., metabolites of zeranol, trenbolone, and melengesterol acetate) and are not approved for use in dairy cows. These compounds would not be expected to be detected in dairy lagoons, manure piles, or application fields associated with dairy operations. However, they can be used in other animals such as beef cattle. If these compounds are detected in water wells, it may be an indication that a source other than dairy cows or humans may be responsible.

#### ~~E.D.~~ Isotopic Analysis

Samples from all the water well, dairy lagoon, and WWTP influent were submitted to the University of Nebraska, Lincoln Laboratory for isotopic analysis. The results of the isotopic analyses are presented in **Appendix A14**. A more detailed discussion regarding the interpretation of the isotopic data can be found in Appendix B.

The isotopic analysis is used to identify the general source, or combination of sources, or dominant processes that have contributed nitrates to the drinking water wells evaluated in this study (Kendall and McDonnell 1998 and Michener and Lajtha 2007). Most of the literature on isotopic fractionation, particularly the part attributing specific samples to specific sources, makes clear that the science is still evolving and that this tool is most appropriately ~~used as a~~ supplement ~~to~~ other methods used to investigate the source of nitrates (Kendall and others 2007).

Isotopes are forms of the same element that have a different number of neutrons. As an example, the atomic weight of nitrogen is 14.0067 because the most common isotope of nitrogen is the form with seven neutrons and seven protons and a mass number of 14, written as  $^{14}\text{N}$ .  $^{14}\text{N}$  makes up 99.636 percent of the total nitrogen in the atmosphere and is referred to as the "light isotope." Nitrogen 15 consists of seven protons and eight neutrons and is written as  $^{15}\text{N}$ .  $^{15}\text{N}$  makes up the rest of the total nitrogen in the atmosphere at 0.364 percent and is referred to as the "heavy" isotope.

Isotopic values are reported as the ratio of the heavy isotope (in this case,  $^{15}\text{N}$ ) to the light isotope (in this case,  $^{14}\text{N}$ ) in the sample compared with that ratio in a chosen standard. For nitrogen, the standard is the pool of nitrogen in the earth's atmosphere ~~or, referred to as~~ the atmospheric standard. Nitrogen isotopic composition is expressed in terms of "delta  $^{15}\text{N}$ ," which is written as  $\delta^{15}\text{N}$  and is expressed as parts per thousand differences from the atmospheric standard stated as, "per mil" or written as ‰.

$$\delta^{15}\text{N} (\text{‰}) = \left( \frac{^{15}\text{N}/^{14}\text{N}}{^{15}\text{N}/^{14}\text{N}} \right)_{\text{sample}} - \left( \frac{^{15}\text{N}/^{14}\text{N}}{^{15}\text{N}/^{14}\text{N}} \right)_{\text{standard}} * 1000$$

[ PAGE \\* MERGEFORMAT ]



$$(^{15}\text{N}/^{14}\text{N})_{\text{standard}}$$

$\delta^{15}\text{N}$  will be positive (e.g., +6.1‰) and therefore heavier if there is more of the  $^{15}\text{N}$  compared with the atmospheric standard in the sample.  $\delta^{15}\text{N}$  will be negative (e.g., -0.2‰), or lighter, if there is less of  $^{15}\text{N}$  in the sample compared with the atmospheric standard.

Isotopes of oxygen ( $^{18}\text{O}$ ) have also been used to provide information on the source of the nitrate in a sample. The standard for  $^{18}\text{O}$  is "Standard Mean Ocean Water," or SMOW. The  $\delta^{18}\text{O}$  in the atmosphere is heavier at 23.5‰. Nitrate derived from atmospheric deposition is therefore heavier with a  $\delta^{18}\text{O}$  of 60‰ to 70‰.

#### G.E. Age Dating and Gas Study

Several methods are available to measure the age of groundwater in a well, meaning the amount of time between the initial infiltration of the water into the ground and the time it was sampled in the well. For this study, EPA selected a method involving sampling for sulfur hexafluoride ( $\text{SF}_6$ ).  $\text{SF}_6$  was selected because some of the wells in this study were anoxic, and  $\text{SF}_6$  is stable in anoxic groundwater.  $\text{SF}_6$  is useful in age-dating because it has been increasing in the atmosphere as it is released by human activities.

$\text{SF}_6$  is a liquid at room temperature and is used in high voltage switches and capacitors as a replacement for polychlorinated biphenyls (PCBs). Significant production of  $\text{SF}_6$  began in the 1960s for use in high-voltage electrical switches.  $\text{SF}_6$  is extremely stable, with an estimated atmospheric lifetime of 800 years (Morris and others 1995) to 3200 years (Ravishankara and others 1993). As more of it is used, more of it escapes into the atmosphere. It is very persistent in the atmosphere, so the concentration has been steadily increasing. Additionally,  $\text{SF}_6$  does not biodegrade even in areas of low oxygen, which is true for some of the Yakima Basin. Due to this increase, laboratory measurements of the concentration in groundwater can indicate the time since the groundwater was last in contact with the atmosphere. Measurement of the concentration in groundwater indicates the time when the sample infiltrated into the ground.

All water wells were sampled for  $\text{SF}_6$ . The analysis was completed by the USGS laboratory in Reston, Virginia.  $\text{SF}_6$  is not an analysis done by commercial laboratories. The Reston laboratory was selected because it had developed a method that had been used successfully by the USGS in Washington State. The purpose of using age dating was to determine the time since infiltration into the water wells and to attempt to evaluate whether the nitrate found in water wells could be associated with either legacy or current practices. A summary of the results for the age dating and gas study are in **Appendix A15**.

In addition to the  $\text{SF}_6$  samples, five gas studies were conducted. These studies involved filling containers with water for the analysis of nitrogen and argon gas to measure the temperature and elevation of the recharge zone for the groundwater. These data are used to correct the  $\text{SF}_6$  measurement for excess nitrogen, which can be dissolved when groundwater elevations fluctuate rapidly. It also provides a means to determine if nitrogen gas has been added to the sample from

**Commented [LE46]:** this confused me when I first read it. I thought "there are easier ways to measure temperature and elevation" before I realized this analysis was intended to figure out where the recharge zone was! So there needs to be some other introductory text here. You are using the nitrogen and argon measurements to characterize the temp and elevation? To infer the temp and elevation? I'm not sure of the best way to say it.

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denitrifying bacteria breaking down nitrate in an anoxic setting. None of the EPA samples showed evidence of denitrification based on measured nitrogen to argon ratios.

**Commented [LE47]:** Another result reported in this section. Seems like it should be all or none.

As seen in **Appendix A15**, there were no reported SF<sub>6</sub> values for WW-01, WW-11, WW-12, WW-23, WW-27, and WW-28. Values were not reported because the concentration of SF<sub>6</sub> in the groundwater exceeded the highest expected concentration based on average atmospheric concentrations of SF<sub>6</sub>. EPA determined as a result that the SF<sub>6</sub> results for those wells were not meaningful. These samples may indicate areas where localized human caused releases of SF<sub>6</sub> occurred. For example, they could include the accidental release during servicing of high voltage equipment or the intentional introduction of SF<sub>6</sub> into water for localized fate and transport studies or for tracing leaking pipes. Alternatively, volcanic rocks can contain more SF<sub>6</sub> than the average atmospheric concentrations and the volcanic terrain and mineralogy of the sediments in the local aquifer may be the source of the SF<sub>6</sub>. Based on the high values of SF<sub>6</sub> observed in the groundwater, the USGS Reston Laboratory believes that the concentrations were from human caused releases and not related to natural levels associated with volcanic regions.

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VIII. QUALITY ASSURANCE AND QUALITY CONTROL

As discussed previously, the project was implemented in three phases. In Phase 1, a GIS screening application was developed and used to identify potential sample locations and sites in the Lower Yakima Valley for Phase 2 sampling and screening. Phase 1 also developed estimates of the relative nitrogen available for application to the land from different sources. Phase 2 and Phase 3 involved sampling and analysis as described in Sections V, VI and VII. A discussion of the quality assurance and quality control (QA/QC) procedures followed in Phase 2 and Phase 3 and a summary of the data validation process conducted by EPA QA chemists is presented in **Appendix C**.

IX. ANALYTICAL RESULTS AND DISCUSSION

This section presents the analytical results for the Phase 3 sampling conducted at the three source areas previously described: the dairies, residential septic systems, and irrigated croplands. The results are presented Sections A through E below present the results from sampling at the five locations identified for this study (See **Figure 11**), including:

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- Haak Dairy (one location)
- Dairy Cluster (one location with six dairies in close proximity)
- Septic Systems Wastes (three locations: Mabton [one site], N. Harrah [one site], and Sunnyside [two sites and three WWTPs (Zillah, Mabton, and Toppenish)])
- Irrigated/fertilized crop fields (three locations: Mabton [three separate crops], N. Harrah [one crop], and Sunnyside [two separate crops])

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Two sites WW-18 and WW-30 that were not apart of the original study design but which were sampled during the study.

Each section listed above contains five subsections to address each of the different compounds or analytical techniques used to evaluate whether there is a link between the high nitrate levels and different sources: general chemistry; microbiology; organic chemicals; isotopic analyses and age dating. In addition, each of the four main sections provides a summary of the results.

#### ⦿ R&M Haak Dairy

The R&M Haak Dairy is located in an agricultural area north of the Yakima River, about four miles north of the city of Sunnyside. It is in the Benton groundwater basin, which includes the communities of Sunnyside, Grandview, Satus, Kiona, Prosser, Mabton and Richland. This Dairy was selected as one of the sampling locations because it is relatively high on the landscape with very few other sources of nitrate above the dairy. The Dairy A ditch runs from north to south through the Dairy. Cow pens, a milking parlor, and three waste lagoons lie west of the ditch. There are several large structures where cows are kept. East of the ditch, a center-pivot irrigation system is installed on a large sprayfield which is used by the Dairy as a disposal location for liquid wastes. The Dairy operator stated that corn and triticales were alternately grown on the sprayfield. See Appendix D for a more detailed description of the Haak Dairy and its operations.

Figure 11 and Figure 12 shows the Phase 3 sample locations associated with the Haak Dairy. The sampling locations include:

- ⦿ One residential drinking water well upgradient of the dairy (WW-01);
- ⦿ One dairy supply well (WW-02);
- ⦿ One dairy manure pile located on the dairy (SO-01);
- ⦿ Two dairy lagoons with three samples collected (LG-01, LG-02, and LG-03). Lagoon samples LG-02 and LG-03 are from the same lagoon;
- ⦿ One application field sample (SO-02) and;
- ⦿ Three downgradient residential drinking water wells (WW-03, WW-04, and WW-05).

#### 1. Haak Dairy: General Chemistry

The four types of general chemistry data collected at the Haak Dairy were nitrate and other forms of nitrogen; major ions; minor ions and trace inorganic elements; and perchlorate. Each of these is discussed below.

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Commented [LE51]: In Figure 11 legend, re-label "soil" as "application field" to be consistent with the other labels, which are locations, not media.

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#### Haak Dairy: Nitrate and Other Forms of Nitrogen

All five water well samples, three dairy lagoon samples, one manure pile sample, and one application field sample were analyzed for several forms of nitrogen. The water wells and lagoons were analyzed for nitrate, nitrate plus nitrite, ammonia or ammonium (if in an aqueous solution), and TKN. The manure samples and the application field samples that were receiving dairy waste were analyzed for extractable nitrate-N (Nitrate-N Solid), extractable ammonia-N (Ammonia-N Solid), and total nitrogen by combustion (Total Nitrogen Solid).

In addition, the total nitrogen in all forms was calculated for each sample and the value presented summed as "Calculated Total Nitrogen." The manure sample, SO-01, had only 22 percent solids and was analyzed for TKN rather than total nitrogen by combustion. For SO-02, the total nitrogen equals the nitrate plus the TKN value. For all other solid samples, the total nitrogen equals the nitrogen by combustion result.

Figure 12 and Table 5 show the concentration of total nitrogen in parts per million at each of these sampling locations. Total nitrogen is the sum of nitrate, nitrite, and TKN. Using total nitrogen values allows a comparison between among different locations.

**Table 5: Haak Dairy – Concentrations of Different Forms of Nitrogen Including Total Nitrogen Values for Water Wells, Lagoons, Manure Piles, and Application Fields**

Location	Nitrate as N (mg/L)	Nitrate + Nitrite as N (mg/L)	Ammonia as N (mg/L)	TKN as N (mg/L)	Calculated Total Nitrogen (mg/L)
Water Wells and Lagoons					
WW-01: Upgradient Well	0.4	0.4	ND	ND	0.4
WW-02: Supply Well	3.1	3.4	ND	ND	3.4
LG-03: Lagoon Influent	NA	ND	920	1200	1200
LG-04: Lagoon Outlet	NA	1.2	1200	1400	1401
LG-05: Lagoon Outlet	NA	1.0	1200	1400	1401
WW-03: Downgradient Well	33.1	35.5	ND	ND	35.5
WW-04: Downgradient Well	51.9	55.0	ND	ND	55.0
WW-05: Downgradient Well	12.8	13.4	ND	ND	13.4
Manure pile					
Location	Ammonia-N Solid (mg/kg)	Nitrate-N solid (mg/kg)	Total Nitrogen Solid (mg/kg)	Calculated Total N (ppm)	
SO-01: Manure	10,100	0.32	29,700 (as TKN)	29,700	
Application Field					
Location	Ammonium as N (mg/kg)	Nitrate + Nitrate as N (mg/kg)	Total Nitrogen Solid (mg/kg)	Calculated Total N (ppm)	
SO-02: Application	4.6	71.7	2760	2760	

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Field				
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NA: Not Analyzed ND: Not Detected

TKN was not detected in the five water wells and therefore the total nitrogen value is the sum of the nitrate plus nitrite concentrations. For WW-01, the total nitrogen value is 0.4 parts per million (ppm) which is within the “background” nitrogen levels. The downgradient water wells that were sampled (WW-03, WW-04 and WW-05) had total nitrogen concentrations of 35.5 ppm, 55.0 ppm and 13.4 ppm, respectively. These levels are well over background nitrogen levels, and the nitrate levels exceed the drinking water MCL of 10 ppm (mg/L)

Nitrate and nitrite were not detected and therefore the total nitrogen values for the lagoons are reflected by the TKN value. The total nitrogen concentrations in LG-01 were 1,200 ppm and in LG-02 and LG-03 was 1,401 ppm. The total nitrogen concentration for the manure sample was 29,700 ppm and the soil sample was 2,760 ppm.

**Figure 12** and **Table 5** show an increase in concentrations of total nitrogen between the upgradient well and the downgradient wells with likely sources, such as dairy lagoons, manure piles, and application fields receiving dairy waste located between these points. The lagoons, manure piles, and application fields from the Haak Dairy are a likely source of the increased nitrogen levels in the downgradient wells. Other sources of nitrogen, such as inorganic fertilizer, may also be contributing to the nitrogen in the downgradient wells. A Washington State Department of Agriculture inspection report indicates the Haak Dairy has utilized inorganic fertilizer on its application fields, in addition to animal wastes (WSDA, 2010).

**Commented [LE52]:** It is a little unusual to see all these sample types plotted on the same graph, but I get what you are trying to show. It would help if the bars were color or pattern-coded to indicate the different media. Maybe upgradient wells, potential sources, and downgradient wells. Not sure how you'd classify the production wells, however.

This comment applies to all similar figures—I think it would help to illustrate the point

Information on the construction and depth of both the upgradient and downgradient wells would be useful to further confirm the hydraulic connection between the upgradient well, dairy lagoons, and downgradient wells. In addition, information on the construction of the dairy lagoons (if they are lined, and if so, with what material) would be useful to determine the extent to which they may be contributing to the increase in nitrogen concentrations. EPA requested this information from the Haak Dairy via a letter, but it declined to provide it.

**Commented [LE53]:** Did they actively decline (say “no”) or did they just not respond.

#### Haak Dairy: Major Ions

All five wells and three dairy lagoons were sampled for analysis of the major ions. **Figure 13** shows the concentrations of six major ions (calcium, chloride, magnesium, potassium, sodium, and sulfate) in the upgradient well, the dairy lagoons, and the downgradient wells. The concentrations of these six ions all show similar patterns of having higher concentrations in the downgradient wells than the upgradient well.

The difference in concentrations from the upgradient well to downgradient wells ranges from up to: a 3-fold increase for potassium; an 8-fold increase for magnesium; a 10-fold increase for calcium; and more than a 30-fold increase for chloride. Chloride is generally accepted as the most conservatively transported ion and therefore an excellent tracer and possible linking

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candidate. "Conservative" in this case means that the negative chloride ion typically flows with the groundwater unchanged. It is unlikely to participate in reactions or be electrically attracted to minerals such as clays making up the aquifer matrix.

One possible explanation for the observed increase in these major ions, especially for the conservatively transported chloride, is that the dairy lagoons are introducing these ions to the groundwater. After this release, the ions are changing the chemistry and producing the observed higher concentrations in the downgradient wells. As with total nitrogen, this is another indicator of the source of nitrate, although information on the construction and depth of the water wells would be helpful to confirm the hydraulic connection along with information on the construction of the lagoons.

#### Haak Dairy: Minor and Trace Inorganic Elements

All five water wells and three dairy lagoons were sampled for analysis of minor and trace inorganic elements. The only metals found in both the water wells and dairy lagoons were barium and zinc (see **Table 6**). Other metals detected in dairy lagoons (chromium, copper, iron, and manganese) were not found in the water wells. The manure and application field samples were not evaluated for minor or trace inorganic elements.

**Table 6: Haak Dairy - Concentrations of Barium and Zinc in Water Wells and Dairy Lagoons (µg/L)**

Location	Barium	Zinc
WW-01 – Upgradient Well	13.5	Not detected
WW-02 – Supply Well	32.7	5.4
LG-01 – Dairy lagoon	297	1790
LG-02 – Dairy lagoon	931	5410
LG-03 – Dairy lagoon	907	5260
WW-03 – Downgradient Well	135	21
WW-04 – Downgradient Well	178	12
WW-05 – Downgradient Well	164	15

There is an increase in the concentrations from the upgradient to the downgradient wells for both barium and zinc, and the concentrations are higher than seen in other studies for surface water in the area. However, this increase may not be solely attributable to the dairy lagoons, given that barium and zinc are both naturally occurring and their concentrations can vary greatly.

As discussed before, the trace inorganic elements tend to react with aquifer materials. As a result, they are difficult to evaluate as indicators of the linkage between likely sources and downgradient wells. However, some of the elements have been successfully used in similar studies (Davis and others 1998) by evaluating the ratios of specific ions such as bromide to chloride. The ratios can provide evidence that a particular nitrate source has altered the groundwater quality.

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EPA looked for ionic ratios between bromide and chloride, but it was not possible to quantify the amount of bromide in the dairy lagoons because of the concentration and complexity of the dairy lagoon samples. Without the quantification of the concentration of bromide, it was not possible to develop the ratio for the lagoons. The use of the ratio as an indicator of linkage to downgradient wells was therefore not possible despite the detection of these ions in most of the water wells.

#### Haak Dairy: Perchlorate

Perchlorate was analyzed only in the water well samples (see **Appendix A6**). The concentrations ranged from 0.14 µg/L (WW-01) to 1.96 µg/L (WW-03). The results for the perchlorate analysis are evaluated together with the isotopic data because perchlorate was used as an indicator of potential accumulation of atmospherically derived nitrate associated with caliche soils. Perchlorate was not evaluated in the dairy lagoon system, but this compound is not expected to persist in the anoxic environment of a dairy lagoon.

#### 10.2 Haak Dairy: Microbiology

All the wells were sampled for analysis of total coliform and *E. coli*. (see **Appendix A7**). One well (WW-04) had a detectable level of total coliform, but *E. coli* was not detected. MST was not completed for the wells as *E. coli* was not detected in any of the wells. The manure piles and application field samples were not analyzed.

Samples from the three dairy lagoons were analyzed for fecal coliform. High concentrations of fecal coliform were found in the dairy lagoons. MST was performed on the samples from the three dairy lagoons. One of the samples (LG-01) indicated a ruminant source, while two of the samples (LG-02 and LG-03) indicated both human and ruminant sources. As stated before, LG-02 and LG-03 are co-located so the same findings for these two dairy lagoons are not surprising. It is unknown why the two dairy lagoons had an indication of human sources though it is possible that the lagoons are impacted by human waste given the Haak Dairy is on a septic system and has several employees.

#### 11.3 Haak Dairy: Organic Compounds

The organic compounds evaluated included pesticides; trace organics; pharmaceuticals, and hormones.

#### Haak Dairy: Pesticides

Atrazine was the only pesticide detected in the water wells. It was detected in the well upgradient of the Haak Dairy (WW-01), the dairy supply well (WW-02), and two wells downgradient from the Haak Dairy (WW-04 and WW-05). The pesticides were not reported for lagoons because of problems with matrix interference.

The concentrations of atrazine were 0.015 µg/L (WW-01); 0.041 µg/L (WW-02); 0.015 µg/L (WW-04); and 0.11 µg/L (WW-05). Atrazine was detected in the application field sample for the Haak Dairy (SO-02) and the other two corn fields sampled in this study (SO-13 and SO-14). Atrazine is a commonly used herbicide for corn fields and is frequently detected in groundwater beneath both urban and agricultural land uses (Barbash and others 1999).

Both grain and silage corn are a significant feedstock in dairy and other cattle livestock operations, and corn has been grown on fields surrounding the water wells in the past. The detection of atrazine in the application field sample and its presence in the downgradient wells is a likely indication that the application field samples are a contributing source. However, its presence in the upgradient well indicates at least one source of atrazine is upgradient of the dairy.

Three pesticides (Dicamba, Dacthal-DCPA, and 2,4-D) were found in the manure pile sample from the Haak Dairy (SO-01). Atrazine was not detected in any manure pile sample. Given that the three compounds detected are common herbicides, a possible source is the feed given to the dairy cows.

Six pesticides (atrazine; 4-nitrophenol; pentachlorophenol; endosulfan sulfate; chlorpyrifos ethyl; and diuron) were found in the application field sample collected adjacent to the Haak Dairy (SO-02). The application field was historically planted in corn.

#### Haak Dairy: Trace Organics

The only trace organic compound detected in the five water wells was bis-(2-ethylethyl)-phthalate (DEHP) in WW-01 (upgradient well) and WW-03 (downgradient well). It was not detected in any of the dairy lagoon samples (see **Appendix A9**). The trace organics were not analyzed in the manure or soil samples.

Other trace organics were detected in the three dairy lagoons but not detected in any of the downgradient wells: LG-01 (10 compounds); LG-02 (12 compounds); and LG-03 (11 compounds). Compounds found in all three dairy lagoons included: fecal indicators (such as 3-beta-coprostanol and 3-methyl-1h-indole); plant sterols (for example, beta-sitosterol, beta-sigmastanol, and cholesterol); and phenol. Phthalates, such as DEHP, are compounds used in the manufacture of plastics to decrease the brittleness of containers and other objects. They are increasingly ubiquitous in the environment and are being widely detected in water wells (EPA 2011(b)). Given their widespread occurrence and detection in water wells, and their absence in the dairy lagoons, it would be difficult to attribute them ~~with to~~ any source from the Haak Dairy at this time.

#### Haak Dairy: Pharmaceuticals

The UNL completed analysis for two lists of compounds in this study referred to as “wastewater pharmaceuticals” and “veterinary pharmaceuticals.” The wastewater pharmaceuticals analyzed in



this study are generally used by humans. Many of “veterinary pharmaceuticals” can be used in both veterinary practice and to treat humans.

There were no detections in the water wells, manure pile sample, or application field sample for any of the wastewater pharmaceuticals. Thiabendazole was detected in one dairy lagoon (LG-01) sample and DEET was detected in one dairy lagoon (LG-03) sample. Thiabendazole is used to treat worm infections in both livestock and humans and can be used as a pesticide (Mayo Clinic 2011). DEET is a common insect repellent.

Three veterinary pharmaceuticals were detected in one or more water wells (tetracycline, chlorotetracycline, and monesin). Several veterinary compounds were detected in the dairy lagoons: LG-01 (eight compounds); LG-02 (nine compounds) and LG-03 (six compounds). Several compounds were also detected in the manure sample (SO-01: four compounds) and application field sample (SO-02: five compounds).

**Table 7** provides the concentrations of the three compounds-veterinary pharmaceuticals detected in the water wells and the concentrations for these compounds in the dairy lagoons, manure sample, and soil sample from the application field associated with the Haak Dairy. **Appendix A11** provides the concentrations of all the pharmaceuticals detected in the dairy lagoons, manure pile sample, and application field sample.

**Table 7: Haak Dairy - Concentrations of Veterinary Pharmaceuticals Detected in Water Wells, Dairy Lagoons, Manure Pile, and Application Field Samples**

Location	Tetracycline	Chlorotetracycline	Monesin
WW-01 – Upgradient Well	ND	ND	<b>0.027</b>
WW-02 – Supply Well	ND	ND	ND
LG-01 – Dairy Lagoon	<b>1.96 (J)</b>	R	<b>44.97</b>
LG-02 – Dairy Lagoon	<b>5.83 (J)</b>	<b>0.067 (J)</b>	<b>1086</b>
LG-03 – Dairy Lagoon	<b>2.88 (J)</b>	ND	<b>420</b>
WW-03 – Downgradient Well	<b>0.041 (J)</b>	ND	ND
WW-04 – Downgradient Well	<b>0.075 (J)</b>	<b>0.049</b>	ND
WW-05 – Downgradient Well	ND	ND	ND
SO-01 – Manure Sample	<b>178</b>	ND	<b>441</b>
SO-02 – Application Field Sample	<b>26.9</b>	<b>45.6</b>	<b>2.9</b>

µg/L for wells/dairy lagoons and µg/g for manure/soil samples

Method Detection Limit = 0.02 µg/L for wells/dairy lagoons and 0.5 ug/g for manure/soil samples.

ND = Not detected

“J” values mean the compound was positively identified, but the associated numerical value is an estimate.

“R” values mean the data is unusable for all purposes because of analytical problems with the sample.

Tetracycline was detected in two of the downgradient wells (WW-03 and WW-04) and in the dairy lagoons, manure sample, and application field samples. This detection provides a good

~~indication indicates~~ that tetracycline is used at the Haak Dairy and suggests the lagoons, manure piles, or application fields could be the source of the tetracycline in the wells, especially given that the concentrations in the dairy lagoons was considerably higher than in the downgradient wells.

**Commented [LE54]:** And the downgradient well concentrations indicate that some of it is moving into ground water. (I'm not sure why some sections include conclusions here and some don't.)

Chlorotetracycline was detected in one downgradient well (WW-04), one dairy lagoon (LG-02), and the application field sample (SO-02). The detection of chlorotetracycline in one lagoon and the application field sample indicates that the Haak Dairy is using chlorotetracycline.

Monesin was detected in the upgradient well (WW-01), dairy lagoons, manure sample, and application field sample, but not in any downgradient wells. The concentrations of monesin seen in the samples indicate that it is used at the Haak Dairy.

As stated before, organic molecules are subject to a number of factors that affect their fate and transport properties and may cause them to travel differently from nitrate in groundwater. As organic molecules, they are much more likely to sorb to materials in the aquifer, which can greatly retard their migration with respect to nitrate. Their concentrations are also much more likely to decrease during migration by microbial degradation.

#### Haak Dairy: Hormones

EPA's Ada laboratory analyzed five hormones in water wells and dairy lagoon samples associated with the Haak Dairy. The laboratory did not analyze the manure pile or application field samples associated with the Haak Dairy because the laboratory specializes in liquid samples and did not have solid extraction techniques developed at the time of the study. However, soil and manure samples were analyzed by UNL and included the five hormones evaluated by the Ada laboratory.

No hormones analyzed by the Ada Laboratory were detected in the water well samples associated with the Haak Dairy; however, three compounds were detected in each of the three dairy lagoons sampled (see **Appendix A12**).

UNL analyzed 18 hormones in water wells, dairy lagoons, manure pile, and application field sample associated with the Haak Dairy, including the same five hormones as Ada. In the water wells, testosterone was the only hormone detected, and it was detected in all five wells, including the upgradient well.

Several hormones were detected in the three dairy lagoons by UNL (**see Appendix A13**), but testosterone was detected in only LG-01. Seven hormones were detected in LG-01, five in LG-02, and four in LG-03. Several hormones were detected in the manure and application field samples, but testosterone was not detected. Six compounds were detected in SO-01 and two in SO-02.

The concentrations of testosterone in the water wells and LG-01 are 21 nanograms per liter (ng/L) (WW-01); 16 ng/L (WW-02); 32 ng/L (LG-01); 9 ng/L (WW-03); 12 ng/L (WW-04); and 7 ng/L (WW-05).

The concentration of testosterone in the upgradient well (WW-01) was greater than in the downgradient wells (WW-03 to WW-05), although the highest concentration was in LG-01. While other hormones were detected in the dairy lagoons, manure pile, and application field samples, they were not found in the water wells. Given that the concentration of testosterone in the upgradient well is greater than the downgradient wells, it is difficult to determine the likely source of the testosterone, although the concentration in one dairy lagoon sample was higher than in the downgradient wells.

#### 1.3.4. Haak Dairy - Isotopic Analyses

**Table 8** provides the results for the isotopic data for water wells for the Haak Dairy, and **Table 9** below provides the results for the isotopic data for the Haak Dairy lagoons. See **Appendix C** for more details on the interpretation of the isotopic data.

Location	Nitrate-N (mg/L)	$\delta^{15}\text{N-NO}_3$ (‰)	Dominant Source	$\delta^{18}\text{O-NO}_3$ (‰)	Overall Assessment
WW-01	0.2	NM	NM	NM	NM
WW-02	3.0	2.7	Soil Cycling	15	Soil Cycling
WW-03	34	2.3	Fertilizer/Animal Waste	29	Fertilizer & Atmospheric & Animal Waste
WW-04	49.9	3.5	Fertilizer/Animal Waste	-4.5	Fertilizer & Animal Waste
WW-05	12.8	9.7	Animal Waste	7.1	Animal Waste

$\delta^{15}\text{N-NO}_3$ . Values less than 2.0 = dominated by fertilizer; values between 2.0 to 8.4 = undetermined mixture of fertilizer and/or animal waste; values greater than 8.4 = dominated by animal.

$\delta^{18}\text{O-NO}_3$ . Values greater than 20 considered strong atmospheric contribution.

WW-01 had insufficient nitrate to allow analysis. The dominant source of the nitrates for the other wells is attributed to either soil nitrogen from plant natural degradation (WW-02); undetermined mixture of fertilizer, animal waste, and atmospheric deposition (WW-03); undetermined mixture of fertilizer and animal waste (WW-04); or animal waste (WW-05).

**Table 9: Isotopic Analysis - Summary Results for Lagoons**

[ PAGE \\* MERGEFORMAT ]

Location	Position in System	Ammonia (mg/L)	$\delta^{15}\text{N-NH}_4$ (‰)	Assessment
LG-01: Haak	Influent	907	3.4	Fresh Animal Waste
LG-02: Haak	Discharge	923	10.1	Volatilized animal waste
LG-03: Haak	Discharge	896	9.9	Volatilized animal waste

The information for the lagoons supports a conclusion that the source of the nitrogen in the dairy lagoons is associated with animal waste. For the three dairy lagoons, LG-01 was the nearest point of entry to the dairy lagoon system. Dairy lagoon samples LG-02 and LG-03 were collected at the end of the system just before it was pumped onto the application fields.

**Commented [LE55]:** So here there are conclusions regarding the sources in the lagoons, but not the groundwater, even though only the 3 downgradient wells were linked to animal waste in Table 8.

These samples are considered co-located and similar values would be expected. The expected trend would be for the isotopic weight fractions to increase in  $\delta^{15}\text{N-NH}_4$  as the ammonia is volatilized in the dairy lagoons farther from the entry point to the dairy lagoon system. This trend would result in larger numbers for the  $\delta^{15}\text{N-NH}_4$  and is seen for the three dairy lagoons at the Haak Dairy (LG-01 3.37‰ compared with 10.07‰ for LG-02).

### 13.5. Haak Dairy: Age Dating

Age dating data were collected for the water wells. Two samples were collected from each well and the values were averaged (see Table 10).

**Table 10: Haak Dairy – Summary of Age Dating Analyses for Water Wells (Years)**

Location	Sample Age	Duplicate Age	Average
WW-01: Upgradient Well	Over Value	Over Value	NA
WW-02: Supply Well	15.8	16.3	16.1
WW-03: Downgradient Well	24.8	25.8	25.3
WW-04: Downgradient Well	21.8	23.3	22.6
WW-05: Downgradient Well	18.3	20.8	19.6

Over Value: These samples contained more  $\text{SF}_6$  than can be explained by equilibrium with modern air.

No values were reported for WW-01 (reported as an over value – see above). The supply well had an average age of 16.1 years and the average age of the downgradient wells ranged from 19.6 to 25.3. Typically, the supply well would be deeper, yielding water from deeper zones in the aquifer, which have traveled farther and are presumably older. In the case of the supply well at the Haak Dairy (WW-02) the age was measured as younger. The younger than expected age could be the case because the supply well is screened in the same shallow zone as the downgradient wells and the time difference in the age may correspond to the travel time between the wells.

**Commented [LE56]:** That doesn't explain it. What is an over-value? How far back can the method go—does this just mean the water is older than that?

#### 14.6. Haak Dairy – Summary of Results for Residential Water Wells

Table 11 provides a summary of the groups of compounds and analytical techniques (general chemistry, organic compounds, and isotopic analyses) that provide the most useful information to address the question of the likely sources of the nitrate for the four residential water wells associated with the Haak Dairy. No conclusions using the microbial data is possible given the three downgradient wells did not exhibit any microbial contamination. In addition, the age dating data do not provide any specific evidence to connect specific sources to high nitrate levels.

Commented [LE57]: Nice summary! This is really helpful.

**Table 11: Haak Dairy – Summary of Results for Residential Water Wells**

Sample Location	General Chemistry	Organic Compounds Detected in Wells	Organic Compounds also Detected in Dairy Sources	Isotopic Analyses
WW-01 Upgradient Well	Nitrate level = 0.4 mg/L  No trends in total nitrogen or major ions as this is an upgradient well	Atrazine, DEHP, testosterone, and monesin.	Not applicable ** because this is an upgradient well	Not measured because lack of nitrate in sample
WW-03 – Down-gradient Well	Nitrate level = 34 mg/L.  Total nitrogen concentrations increased substantially between WW-01 and WW-03  Ten to 45-fold increase in concentration in four major ions between WW-01 to WW-03.	Atrazine,  DEHP  Tetracycline   Testosterone,	Atrazine (SO-01)  DEHP (ND)  Tetracycline (LG-01, LG-02, LG-03, SO-01, and SO-02)  Testosterone (LG-01)	Fertilizer & Animal & Atmospheric
WW-04 – Down-gradient Well	Nitrate level = 49.9 mg/L  Total nitrogen concentrations increased substantially between WW-01 and WW-04  Ten to 20-fold increase in concentration in four major ions between WW-01 and WW-04.	Atrazine  Tetracycline   Chlorotetracycline  Testosterone	Atrazine (SO-01)  Tetracycline (LG-01, LG-02, LG-03, SO-01, and SO-02)  Chlorotetracycline (LG-02 and SO-02)  Testosterone (LG-01)	Fertilizer & Animal Waste

Sample Location	General Chemistry	Organic Compounds Detected in Wells	Organic Compounds also Detected in Dairy Sources	Isotopic Analyses
WW-05 – Down-gradient Well	<p>Nitrate level = 12.8 mg/L</p> <p>Total nitrogen concentrations increased substantially between WW-01 and WW-05</p> <p>Five to 10-fold increase in concentrations in four major ions between WW-01 and WW-05.</p>	<p>Atrazine</p> <p>Testosterone</p>	<p>Atrazine (SO-01)</p> <p>Testosterone (LG-01)</p>	Animal waste

ND = No Detects

All of the residential water wells, except WW-01 have nitrate levels greater than EPA's MCL of 10 mg/ L. In two cases, the concentrations are substantially greater than the MCL (WW-03 and WW-04). The total nitrogen and major ions data indicated that the dairies are likely contributing to higher levels of nitrogen and major ions in the three downgradient wells associated from the Haak Dairy. The total nitrogen and major ion data indicate an increasing trend in the concentrations from upgradient to downgradient wells, with higher concentrations in the dairy lagoons, manure samples, and application field samples.

Atrazine was the only pesticide detected in the water well samples for the organic chemicals. Atrazine is widely used throughout the area, and the source is likely historical and current use of the pesticide. DEHP was the only trace organic detected in the water wells, but it was not detected in any of the dairy lagoons.

Tetracycline was detected in two of the three downgradient wells and in all three dairy lagoons and the manure and application field samples. It is possible that the source of the tetracycline is the various upgradient sources from the Haak Dairy or it could be septic systems, given tetracycline is used by humans. Monesin was detected in the upgradient well but not in any of the downgradient wells. Monesin was detected in all the dairy lagoons and manure and application field samples.

Testosterone was the only hormone detected in water wells, with higher concentrations in the upgradient well than the downgradient wells; the sources could be from any animal.

It appears that the source of nitrate for each of the wells is different for the isotopic data. For WW-02, the dominant source appears to be soil cycling, while the possible sources in WW-03 are fertilizer, animal waste, and atmospheric. For WW-04, the possible sources are fertilizer and animal waste, while the dominant source for WW-05 is animal waste.

**Commented [LE58]:** This well really doesn't look to be downgradient of many of the dairy sources—is it? If not, it would make sense that it looks different.

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In conclusion, all of the residential water wells except WW-01 have high nitrate levels. Data from the total nitrogen and major ion data indicate an increasing trend in the concentrations from the upgradient well to the downgradient wells. Information on the construction and depth of the wells would be helpful to confirm the contributions of sources to the higher concentrations seen from the upgradient well to the downgradient wells. The tetracycline in the wells could be a contributing source related to the Haak Dairy, but could also be from a human source. The isotopic data provide good-strong evidence that animal waste is a dominant contributor to the nitrate contamination for WW-05.

### **1.3 Dairy Cluster**

The "Dairy Cluster" refers to a group of dairies in close proximity to each other. EPA regards them as three facilities because some of the dairies are adjacent and have common ownership. The Liberty Dairy and the Hank Bosma Dairy are regarded by EPA as a single facility, as are the Cow Palace 1&2 Dairies, and the George DeRuyter/D&A Farms Dairies. Together they occupy about eight square miles of land (roughly 5,100 acres) north of the Yakima River and the town of Liberty, near the northern edge of the irrigated area in the Yakima Valley.

For this study sampling was organized into four areas: (1) George DeRuyter Dairy; (2) D&A Farms; (3) Cow Palace #1 and Cow Palace #2; and (4) Henry Bosma and Liberty Dairy. Appendix E provides a more detailed description of the Dairy Cluster and their operations.

Figure 11 show the sample locations for the Dairy Cluster. The sampling locations include:

- One upgradient drinking water well (WW-06) located north of all the other samples in the Dairy Cluster, with the exception of SO-05;
- Three dairy supply wells located on each of the dairies except for Henry Bosma and Liberty Dairies. The supply well at the Bosma dairy was not sampled because they used an ion-exchange system which modified the water chemistry of the sample and made it unsuitable for sampling for this study. A residential water well owned by the dairy was sampled instead (WW-10). The three dairy supply wells included: George DeRuyter Dairy (WW-07); D&A Farms (WW-08); and Cow Palace #1 and #2 (WW-09);
- Four dairy manure pile samples located on each dairy: George DeRuyter (SO-03), D&A Farms (SO-05); Cow Palace #1 and #2 (SO-07); and Henry Bosma and Liberty Dairy (SO-09);
- Twelve dairy lagoon samples;
  - George DeRuyter (LG-04, LG-05, and LG-06. LG-05 and LG-06 were taken from the same lagoon);

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**Commented [LE59]:** Add explanation that they are generally discussed as a group because they are so close—can't analyze the spatial patterns separately (if that's the reason)

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- D&A Farms (LG-07, LG-08, and LG-09. LG-08 and LG-09 were taken from the same lagoon);
- Cow Palace #1 and #2 (LG-10, LG-11, and LG-12). LG-11 and LG-12 were taken from the same lagoon); and
- Henry Bosma and Liberty Dairy (LG-13, LG-14, and LG-15) (these were three separate lagoons);
- Four dairy application field samples;
  - George DeRuyter (SO-04);
  - D&A Farms (SO-06);
  - Cow Palace #1 and #2 (SO-08); and
  - Henry Bosma and Liberty Dairy (SO-10).
- Eight downgradient residential drinking water wells (WW-10 to WW-17)

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#### Dairy Cluster: General Chemistry

The four types of general chemistry data collected at the Dairy Cluster were: nitrate and other forms of nitrogen; major ions; minor ions and trace inorganic elements; and perchlorate. Each of these is discussed below.

#### Dairy Cluster: Nitrate and other forms of nitrogen

Twelve water wells and 12 dairy lagoon samples were analyzed for nitrate, nitrate plus nitrite, ammonia or ammonium, and TKN. The manure samples and agricultural field samples that were receiving dairy waste were analyzed for extractable nitrate-N (Nitrate-N Solid), extractable ammonia-N (Ammonia-N Solid), and total nitrogen by combustion (Total Nitrogen Solid). In addition, total nitrogen from all forms was calculated for each sample and is present as "Calculated Total Nitrogen." **Table 12** shows the values measured for total nitrogen for all the dairy cluster samples.

**Table 12: Dairy Cluster - Concentrations of Forms of Nitrogen Including Total Nitrogen Values for Water Wells, Lagoons, Manure Piles, and Application Fields**

Location	Nitrate as N (mg/L)	Nitrate + Nitrite as N (mg/L)	Ammonia as N (mg/L)	TKN as N (mg/L)	Total Nitrogen (mg/L)
<b>Water Wells and Lagoons</b>					
WW-06: Upgradient Well	0.71	0.73	ND	ND	0.73

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WW-07: Supply Well	1.02	1.19	ND	ND	1.19
WW-08: Supply Well	11.7	12.9	ND	ND	12.9
WW-09: Supply Well	ND	ND	ND	ND	ND
LG-04: Lagoon Influent	ND	ND	920	1600	1600
LG-05: Lagoon Outlet	ND	ND	1200	1600	1600
LG-06: Lagoon Outlet	ND	3.1	1200	1800	1803
LG-07: Lagoon Influent	3.1	3.1	950	1700	1703
LG-08: Lagoon Outlet	ND	ND	730	1200	1200
LG-09: Lagoon Outlet	ND	ND	760	1100	1100
LG-10: Lagoon Influent	ND	ND	190	380	380
LG-11: Lagoon Outlet	ND	ND	240	500	500
LG-12: Lagoon Outlet	ND	ND	240	290	290
LG-13: Lagoon Influent	2.5	2.5	970	1700	1703
LG-14: Lagoon Outlet	ND	ND	860	1400	1400
LG-15: Lagoon Outlet	ND	ND	560	900	900
WW-10: Downgradient Well	ND	ND	ND	ND	ND
WW-11: Downgradient Well	22.3	23	ND	ND	23
WW-12: Downgradient Well	45	46.7	ND	ND	46.7
WW-13: Downgradient Well	41.4	44	ND	ND	44
WW-14: Downgradient Well	40.9	43.4	ND	ND	43.4
WW-15: Downgradient Well	29.4	30.2	ND	ND	30.2
WW-16: Downgradient Well	22.3	23.4	ND	ND	23.4
WW-17: Downgradient Well	21.7	22.7	ND	ND	22.7
Manure Piles					
Location	Ammonia-N Solid (mg/kg)	Nitrate-N solid (mg/kg)	Total Nitrogen Solid (mg/kg)	Calculated Total N (ppm)	
SO-03: Manure	1470	32.8	9210	9210	
SO-05: Manure	1060	43.1	13600	13600	
SO-07: Manure	3600	18.9	16100	16100	
SO-09: Manure	1700	5.69	13700	13700	
Application Fields					
Location	Ammonium as N (mg/kg)	Nitrate + Nitrate as N (mg/kg)	Total Nitrogen Solid (mg/kg)	Calculated Total N (ppm)	
SO-04: Application field	7.3	247	2110	2110	
SO-06: Application field	6.8	45.6	960	960	
SO-08: Application field	2.9	84.3	3040	3040	
SO-10: Application field	7.1	139	3590	3590	

Figures 14a, 14b, 14c, and 14d shows the concentration of total nitrogen for the DeRuyter Dairy, D&A Dairy, Cow Palace # 1 and #2 Dairy, and Bosma and Liberty Dairy. As with the Haak Dairy, there is an increasing trend in concentration of total nitrogen from the upgradient

well to the downgradient wells, with several likely sources of nitrogen in between (dairy lagoons, manure piles, and application fields). The attribution to a specific source is complicated given the lack of information on water wells, but it does suggest the lagoons, manure piles, or application fields are a likely source contributing to the higher total nitrogen concentrations observed downgradient from these sources.

#### Dairy Cluster: Major Ions

Figures 15a, 15b, and 15c show the concentrations of several major ion in the upgradient water wells, the supply wells, the lagoons, and the downgradient wells. An average concentration for the lagoons for each area was calculated: LG-04, LG-05, and LG-06; LG-07, LG-08, and LG-09; LG-10, LG-11, and LG-12; and LG-13, LG-14, and LG-15. The averages for the lagoons were calculated for each area because they are in close proximity and they allow easier comparison.

The figures show a similar pattern to that observed at the Haak Dairy of elevated concentrations in the downgradient wells (WW-10 to WW-17) compared with the upgradient wells (WW-06) and supply wells (WW-07 to WW-09). The increase in the concentrations ranges from up to: seven-fold for sodium; nine-fold for magnesium; ten-fold for calcium; and almost 40-fold increase for chloride. Potassium did not show any clear increase.

Sulfate ~~saw showed~~ a large increase in concentration in the downgradient wells compared to the upgradient well, but also ~~saw showed~~ an increase in the concentrations ~~from compared to the~~ lagoons. The reason for this pattern is that ~~similar to as with~~ nitrogen, sulfur comes in many different forms. This study only evaluated sulfate, which is the oxidized form of sulfur. The levels of sulfate in the lagoons is less than in the downgradient wells because the form of sulfur in the lagoons is sulfide. The sulfide to sulfate transformation occurs outside the lagoon when it is exposed to oxygen. This accounts for the higher levels of sulfate in the downgradient wells.

Commented [LE60]: If that's what you mean

As with the Haak Dairy, one possible explanation for the observed increase in these major ions, especially for the conservatively transported chloride, is that the dairy lagoons are introducing these ions to the groundwater. After the release, the ions are then changing the ground water chemistry and producing the observed higher concentrations in the downgradient wells.

#### Dairy Cluster: Minor and Trace Inorganic Elements

All water wells and dairy lagoons were sampled for analysis of minor and trace inorganic elements (see Appendix A5). The manure and application field samples were not evaluated for minor or trace inorganic elements. The trace inorganic elements found in the water wells and dairy lagoons were barium, iron, manganese, mercury, and zinc. Barium was detected in all 11 wells, iron was detected in five wells, manganese was detected in four wells, mercury was detected in one well, and zinc was detected in eight wells. No discernible pattern of increasing concentrations from the upgradient wells to the downgradient wells was evident for any of these compounds with the possible exception of barium. This lack of a pattern is not surprising, since

minor ions and trace inorganic elements are not generally used as linking compounds because they are ubiquitous and are frequently not conservatively transported with groundwater.

#### Dairy Cluster: Perchlorate

Perchlorate analysis was performed on all the water well samples (see **Appendix A6**). The concentrations ranged from less than the detection limit (0.003 µg/L) to 3.08 µg/L (WW-17). Perchlorate was intended to augment the isotopic data as an indicator of potential accumulation of atmospherically derived nitrate associated with caliche soils. However, elevated levels of perchlorate were seen in only two wells. None of those wells was part of the Dairy Cluster. Perchlorate was not evaluated in the dairy lagoon system because this compound is rapidly degraded in the anoxic environment of a dairy lagoon.

### **1. Dairy Cluster: Microbiology**

There were no detections of total coliform, fecal coliform, or *E.coli* at any of the water wells either upgradient of the Dairy Cluster, in the supply wells, or downgradient of the Dairy Cluster. MST was not performed because there was no indication of fecal contamination.

All the dairy lagoons in the Dairy Cluster were analyzed for fecal coliform. LG-04 through LG-09 were also analyzed for *E.coli* and MST was performed. The other dairy lagoons did not have *E. coli* or MST performed because the Manchester mobile laboratory was only able to participate in the sampling effort for a limited period (see **Appendix A7**).

All the dairy lagoons had high levels of fecal coliform. Of the six dairy lagoons evaluated using MST, five indicated a ruminant source (LG-04, LG-05, LG-06, LG-07, and LG-08) while one indicated both a ruminant and a human source (LG-09). The five dairy lagoons indicative of ruminant sources are expected, but LG-09 indicated both a ruminant and a human source and is unexpected. It is unknown why LG-09 had an indication of human sources though it is possible that the lagoons are impacted by human waste given the Dairy is on septic systems and they have several employees.

### **1.5. Dairy Cluster: Organic Compounds**

#### Dairy Cluster: Pesticides

Four pesticides were detected in the water wells associated with the Dairy Cluster (atrazine, bentazon, alachlor, and ioxynil). Atrazine, bentazon, and alachlor are all common pesticides used in agricultural production. Ioxynil is not registered for use in the United States (PAN 2011).

- Atrazine: WW-12, WW-13, WW-14, WW-15, WW-16, and WW-17
- Bentazon: WW-08 (supply well)
- Alachlor: WW-13 and WW-17
- Ioxynil: WW-13

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As stated before, there are no results for pesticides in dairy lagoons or WWTPs because of problems with matrix interference from the wastes. The four pesticides detected in the water wells were not detected in the manure or application field samples. The concentrations of atrazine found in the water wells ranged from 0.016 µg/L to 0.18 µg/L.

The concentration of bentazon in WW-08 was 0.036 µg/L. Alachlor levels found in the water wells were 0.048 µg/L and 0.057 µg/L. The concentration of Ioxynil in this study was 0.063.ug/L. The four pesticides are not anticipated to be used in animal operations at the dairies for pest control (Pike 2004), but atrazine, alachlor, and bentazon may be used in corn fields that produce grain for dairy feedstock. Each of the dairies includes crop land where pesticides may have been applied. Given the historical use of these pesticides and the detection of these compounds in other studies, it is likely that these pesticides are from the current and historical use of pesticides for agricultural purposes, which could include application by the dairies.

Seven pesticides were detected in one of more of the manure samples. These pesticides were not detected in the water well samples. One possible source of these pesticides is from the feed given to the animals. Seven pesticides were also detected in one or more of the field application samples, but they were not detected in the water well samples.

#### Dairy Cluster: Trace organics

For the trace organics, three compounds were detected in water well samples associated with the dairy cluster:

- Bis-(2-ethylhexyl)-phthalate (DEHP) in WW-06 (upgradient well), WW-11, and WW-17
- Naphthalene in WW-07 (supply well)
- Tetrachloroethylene in WW-07 (supply well).

All 12 dairy lagoons associated with the dairy cluster had one or more detections for trace organics (see **Appendix A9**). Eight compounds were detected in all 12 dairy lagoons associated with the dairy cluster. These compounds are generally the same as were detected in at the Haak Dairy: 3-beta-coprostanol; 3-methyl-ih-indole (skatol); 4-nonyphenol monoethoxylate; beta-sitosterol; beta-stigmastanol; cholesterol; p-cresol; and phenol. Trace organics were not analyzed in manure or soil samples.

Of the three compounds found in water wells, only DEHP was found in one dairy lagoon sample (LG-10). Naphthalene and tetrachloroethylene were not detected in any of the dairy lagoons. The source for these compounds could be septic systems or some other source, given their common usage. DEHP is a common plasticizer and it could come from multiple sources.

#### Dairy Cluster: Pharmaceuticals

DEET was the only wastewater pharmaceutical detected in one ~~supply~~ well (WW-10 – downgradient well). There were no detections of any of the wastewater pharmaceuticals in the manure pile or field application samples.

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Three wastewater pharmaceutical compounds were detected in dairy lagoons associated with the Dairy Cluster: DEET (eight dairy lagoons); diphenhydramine (two dairy lagoons); and thiabendazole (three dairy lagoons). The source of the DEET could be its use as an insect repellent. The source of the diphenhydramine in the dairy lagoons is unknown. Diphenhydramine is a common antihistamine used by humans and can be used in dogs and cats. Thiabendazole is a parasiticide that is used to treat worm infections in both livestock and humans and can be used as a pesticide (Mayo Clinic 2011). It is possible that the source of thiabendazole is its use to treat worm infections.

Eight compounds were detected in water wells with five detected in downgradient wells for the veterinary pharmaceuticals (see **Table 13 - attached**). Three of the eight compounds were detected only in supply wells (erythromycin, sulfadimethoxine, and sulfamethazine). Three residential wells had no detections (WW-12, WW-15, and WW-16). Veterinary pharmaceuticals were detected in the following residential water wells.

Chlorotetracycline: WW-13 and WW-14

Monesin: WW-14

Tetracycline: WW-11, and WW-17

Tylosin: WW-11

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#### Virginiamycin: WW-13

Several of the veterinary pharmaceuticals were detected in the majority of dairy lagoons, manure samples, and application field samples (ractopamine, sulfachloropyridazine, sulfadimethoxine, sulfamethazine, and sulfathiazole) but were not detected in downgradient water wells. The presence of these compounds in the dairy lagoons, manure samples, and application field samples indicates these compounds are used at the dairies but were not transported or were not transported at detectable levels to the downgradient wells during the study period..

The concentration of chlorotetracycline in the downgradient wells was greater than the concentrations in the two dairy lagoons with detections of this compound (**Table 13 – attached**). The two dairy lagoons (LG-05 and LG-10) with detected levels of chlorotetracycline are a considerable distance from WW-13 and WW-14. Chlorotetracycline was detected in the majority of the manure and application field samples, although some of these samples are a considerable distance from WW-13 and WW-14 (such as SO-07 and SO-05). It is possible the source of the chlorotetracycline in the water wells could be the manure piles or application fields.

Monesin was detected in WW-14 and was also detected in high concentrations in the dairy lagoons, manure piles, and application field samples. These detections ~~would~~ indicate that monesin is used at the dairies. Monesin is not used by humans, but is used in dairy cows. Thus, the source of monesin in WW-14 is probably from its use in one or more of the upgradient dairies. This is reinforced by the isotopic findings, which indicate that the source of nitrate for WW-14 is animal waste (although animal waste ~~can be~~ includes human sources).

Tetracycline was detected in upgradient well WW-06 (0.051 µg/L) and in two downgradient wells, which had lower concentrations than the upgradient well (0.038 µg/L for WW-11; and 0.049 µg/L for WW-17). Tetracycline was detected in all of the dairy lagoon samples, manure samples, and application field samples, indicating that tetracycline is used at the dairies. It is possible that tetracycline in the downgradient wells was from one of the sources in the dairy cluster.

Tylosin was detected in one downgradient well (WW-11) and five of the dairy lagoons along with several manure and application field samples. These detections indicate that tylosin is used at the dairies. Similar to monesin, this compound is not approved for use in humans, but it can be used in different livestock, including dairy cattle.

#### Dairy Cluster: Hormones

EPA's Ada laboratory analyzed for five hormones in water wells and dairy lagoons associated with the dairy cluster. The laboratory did not analyze the manure pile or application fields samples associated with the Dairy Cluster. The Ada laboratory did not detect any of the five hormones in water wells associated with the Dairy Cluster. The 12 dairy lagoons all had detected levels of 17- $\alpha$ -estradiol, 17- $\beta$ -estradiol, and estrone, while 17- $\alpha$ -ethyl-estradiol and estriol were not detected in any of the dairy lagoons (**see Appendix A11**).

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The UNL analyzed 18 hormones in water well, dairy lagoon, manure pile, and application field samples associated with the Dairy Cluster, including the same five hormones as Ada analyzed. **Table 14** (attached) provides the results of these analyses for those hormones detected in water wells and also in either the dairy lagoons, manure piles, or application field samples associated with Dairy Cluster.

Hormones were not detected in water wells WW-10, WW-13, WW-14, and WW-16. Nine hormones from the UNL analysis were detected in water wells (see **Table 14 - attached**) with three detected in downgradient wells (WW-11, WW-12, WW-15, and WW-17).

- \*- Androsterone: WW-12, WW-15, and WW-17
- \*- Androstenedione: WW-12.
- \*- Testosterone: WW-11.

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The six compounds detected in dairy supply wells, but not the downgradient wells, include  $\alpha$ -estradiol; 17- $\beta$ -estradiol; 17- $\beta$ -trenbolone;  $\alpha$ -zearalanol; epitestosterone; and 11-keto-testosterone. The following is a discussion on the likely sources for the three compounds detected in the downgradient wells.

Androsterone was not detected in any of the dairy lagoons, manure, or application field samples. The source of the androsterone is unknown. Androsterone is a metabolite of testosterone, so it could come from either humans or other animals or from the dairy lagoons since testosterone was detected in the dairy lagoons.

Androstenedione was detected in three dairy lagoons and all of the manure samples, indicating its likely usage at the dairies. The three dairy lagoons (LG-05, LG-11, and LG-12) with detections of the compounds are a considerable distance from WW-12.

Androstenedione is a precursor to boldenone (a synthetic growth promoter) and is used mainly to treat horses and cattle.

Testosterone was detected in nine dairy lagoons and one manure sample (SO-03). The detection of testosterone in the dairy lagoons is not surprising given it is a natural sex hormone. The source of the testosterone in WW-11 could be the Dairy Cluster.

#### **16.3. Dairy Cluster: Isotopic Analysis**

Isotopic analyses were completed for WW-06 to WW-17 (see **Table 15 -below**). There was insufficient nitrate in WW-06, WW-09, and WW-10 to complete the analysis. Additional details on the results of isotopic analyses conducted for this study are provided in Appendix B of this report.

**Table 15: Dairy Cluster – Summary of Isotopic Analysis for Water Wells**

Location	Nitrate -N (mg/L)	$\delta^{15}\text{N}$ - NO3 (‰)	Dominant Source	$\delta^{18}\text{O}$ -NO3 (‰)	Overall Assessment
WW-06	0.6	NM	NM	NM	NM
WW-07	1.1	-0.1	Fertilizer	NM	Fertilizer
WW-08	11.7	5.3	Fertilizer & Animal Waste	23	Fertilizer & Atmospheric & Animal Waste
WW-09	NM	NM	NM	NM	NM
WW-10	NM	NM	NM	NM	NM
WW-11	21.6	3.0	Fertilizer & Animal Waste	18	Fertilizer & Animal Waste
WW-12	43.6	6.2	Fertilizer & Animal Waste	-1.4	Fertilizer & Animal Waste
WW-13	42	11	Animal Waste	16	Animal Waste
WW-14	40.7	10	Animal Waste	8.5	Animal Waste
WW-15	27.4	5.2	Fertilizer & Animal Waste	30	Fertilizer & Atmospheric & Animal Waste
WW-16	23	5.9	Fertilizer & Animal Waste	5.8	Fertilizer & Animal Waste
WW-17	23.3	6.9	Fertilizer & Animal Waste	2.5	Fertilizer & Animal Waste

$\delta^{15}\text{N}$ -NO3. Values less than 2.0 = dominated by fertilizer; values between 2.0 to 8.4 = undetermined mixture of fertilizer and/or animal waste; values greater than 8.4 = dominated by animal.

$\delta^{18}\text{O}$ -NO3. Values greater than 20 considered strong atmospheric contribution.

The dominant source of nitrate for two wells (WW-13 and WW-14) is animal waste. The dominant source of nitrate for one well (WW-07) is fertilizer. For the other wells, it is not possible to determine the dominant source. For WW-08 and WW-15 the atmospheric contribution is strong.

**Table 16: Isotopic Analysis - Summary Results for Lagoons**

Location	Position in System	Ammonia (mg/L)	$\delta^{15}\text{N}$ -NH4 (‰)	Assessment
LG-04: DeRuyter	Influent	899	6.7	Volatilization Animal Waste
LG-05: DeRuyter	Discharge	1151	10.6	Volatilization Animal Waste
LG-06: DeRuyter	Discharge	1293	10.3	Volatilization Animal Waste
LG-07: D&A	Influent	869	5.4	Fresh Animal Waste
LG-08: D&A	Discharge	696	10.3	Volatilization Animal Waste

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LG-09: D&A	Discharge	658	10.1	Volatilization Animal Waste
LG-10: Cow Palace	Influent	NM	NM	NM
LG-11: Cow Palace	Discharge	274	3.1	Fresh Animal Waste
LG-12: Cow Palace	Discharge	222	2.0	Fresh Animal Waste
LG-13: Bosma	Influent	469	4.4	Fresh Animal Waste
LG-14: Bosma	Discharge	600	3.3	Fresh Animal Waste
LG-15: Bosma	Discharge	658	13.9	Volatilization Animal Waste

Isotopic analyses were completed for the dairy lagoon samples (LG-04 to LG-15). **Table 16** indicates the source of nitrate in dairy lagoons is animal waste. Additional details on the results of isotopic analyses conducted for this study are provided in Appendix B of this report.

#### 17.4. Dairy Cluster: Age Dating

**Table 17** presents the age dating data, and similar to the Haak Dairy, two samples were collected for each water well.

**Table 17: Dairy Cluster – Summary of Age Dating for Water Wells (Years)**

Location	Sample Age	Duplicate Age	Average of Samples	Average of Group
WW-06I	16.3	15.8	16.1	16.1
WW-07	36.3	32.8	34.6	42.5
WW-08	35.3	40.8	38.1	
WW-09	58.3	51.3	54.8	
WW-10	44.3	44.8	44.6	
WW-11	Over Value	Over Value	NA	34.2
WW-12	Over Value	Over Value	NA	
WW-13	24.3	23.8	24.1	
WW-14	30.8	29.3	45.5	
WW-15	27.8	28.3	28.1	
WW-16	29.8	28.8	29.3	
WW-17I	33.3	33.8	33.6	

Over Value: These samples contained more SF<sub>6</sub> than can be explained by equilibrium with modern air.

For evaluation, averages were calculated for the upgradient well (WW-06), three supply wells (WW-07, WW-08, and WW-09), and the six downgradient wells with reported values (WW-10, WW-13, WW-14, WW-15, WW-16, and WW-17). The results indicate the “youngest” water was sampled in the upgradient well, with an average age of 16.1 years. The “oldest” waters were in supply wells associated with the Dairy Cluster, with an average age of 42.5 years. The average age of the waters in the downgradient wells was 34.2.

18.5. Dairy Cluster – Summary of Results for Residential Water Wells

Table 18 provides a summary of the groups of compounds (general chemistry and organic compounds) and analytical techniques (isotopic analyses) that provide information useful to address the question of the likely sources of the nitrate for the nine residential water wells associated with the dairy cluster. No conclusions using the microbial data is possible given the downgradient wells did not exhibit any microbial contamination. In addition, the age dating data do not provide any specific evidence to connect specific sources to high nitrate levels.

Table 18: Dairy Cluster – Summary of Results for Residential Water Wells

WW-06 – Upgradient Well			
General Chemistry	Organic Compounds Detected in Water Wells	Organic Compounds Also Detected in Dairy Sources	Isotopic Analyses
Nitrate level = 0.6 mg/L  No trends in total nitrogen or major ions as upgradient well	Atrazine and DEHP	No comparison because upgradient well	Not sufficient nitrate
WW-10 – Downgradient Well			
Nitrate level = Not detected  No large trends in total nitrogen or major ions between WW-06 and WW-10	DEET  Monesin	DEET (8 lagoons)  Monensin (All the dairy sources except LG-07)	Not sufficient nitrate
WW-11 – Downgradient Well			
Nitrate levels = 21.6 mg/L  Total nitrogen increased 20-fold between WW-06 and WW-11  Three to 25-fold increase in concentration of five major ions between WW-06 and WW-11.	DEHP  Tetracycline  Tylosin	DEHP (LG-10)  Tetracycline (All dairy sources)  Tylosin (5 lagoons, 2 manure samples, and one application field sample)	Fertilizer & Animal Waste
WW-12 – Downgradient Well			
Nitrate level = 43.6 mg/L  Total nitrogen increased 50-fold between WW-06 and WW-12  Five to 25-fold increase in concentration of five major ions between WW-06 and WW-12.	Atrazine  Androstenedienedione  Androsterone	Atrazine (ND)  Androstenedienedione (three lagoons and 4 manure samples)  Androsterone (ND)	Fertilizer & Animal Waste

WW-13 – Downgradient Well			
Nitrate level = 42.0 mg/L	Alachlor	Alachlor (ND)	Animal waste
Total nitrogen increased 40-fold between WW-06 and WW-13	Atrazine	Atrazine (ND)	
Seven to 40-fold increase in concentration of five major ions between WW-06 and WW-13.	Ioxynil	Ioxynil (ND)	
	Virginiamycin	Virginiamycin (5 lagoons)	
WW-14 – Downgradient Well			
Nitrate level = 40.7 mg/L	Atrazine	Atrazine (ND)	Animal waste
Total nitrogen increased 40-fold between WW-06 and WW-14	Monesin	Monesin (All the dairy sources except LG-07)	
Two to 50-fold increase in concentration of six major ions between WW-06 and WW-14.			
WW-15 – Downgradient Well			
Nitrate level = 27.4 mg/L	Atrazine	Atrazine (ND)	Fertilizer & Animal Waster& atmospheric
Total nitrogen increased 30-fold between WW-06 and WW-15	Chlorotetracycline	Chlorotetracycline (LG-10, all manure and application fields except SO-09 and SO-10)	
Two to 20-fold increase in concentration of six major ions between WW-06 and WW-15.	Androsterone	Androsterone (ND)	
WW-16 – Downgradient Well			
Nitrate level = 23.0 mg/L	Atrazine	Atrazine (ND)	Fertilizer & animal waste
Total nitrogen increased 20-fold between WW-06 and WW-16			
Four to 30-fold increase in concentration of five major ions between WW-06 and WW-16.			

WW-17 – Downgradient Well			
Nitrate level = 23.3 mg/L  Total nitrogen increased 20-fold between WW-06 and WW-11  Four to 30-fold increase in concentration of five major ions between WW-06 and WW-17.	Alachlor	Alachlor (ND)	Fertilizer & Animal waste
	Atrazine	Atrazine (ND)	
	DEHP	DEHP (LG-10)	
	Tetracycline	Tetracycline (All dairy sources)	
	Androsterone	Androsterone (ND)	

ND = Not Detected

As with the Haak Dairy, the nitrate levels in the downgradient residential wells are substantially greater than EPA's MCL for nitrate with the exception of WW-06 and WW-10. The total nitrogen and major ions data show increasing concentrations from the upgradient well, WW-06, past several sources of nitrogen, and to the downgradient wells. The downgradient wells contain substantially more nitrogen than is present in the upgradient well (or in some wells elsewhere in the this area such as WW-09 and WW-10). The upgradient well, WW-06, shows low nitrate levels within "background" range. The age of the water in WW-10 is significantly greater than the other downgradient wells, suggesting ~~this~~ this well may be substantially deeper than the other residential wells and less exposed to contamination from the shallow aquifer.

The major ion data, especially for calcium and chloride, show an increasing trend between the dairy waste (dairy lagoons, manure piles, and application fields) and the downgradient wells with high nitrate.

Four pesticides were detected in the water wells associated with the Dairy Cluster. However, none of these pesticides was detected in the manure or application field samples. Because of problems with matrix interference, no results for pesticides in dairy lagoons or WWTPs influent are available. Three trace organics were detected in water wells, but only one (DEHP) was detected in a downgradient well (WW-17). The pesticides data indicate that the source is probably the past or current application for agriculture.

Pharmaceuticals were detected in four downgradient wells. Several of the compounds — tetracycline (WW-11 and WW17); oxytetracycline (WW13 and WW-14); and monesin (W-14) — were detected in dairy lagoons, manure samples, and application fields associated with the dairy cluster. It is possible that the source of the three compounds is one of the Dairy Cluster sources, especially likely for monesin, which is used in dairy cows but not by humans.

Nine hormones were detected in water wells associated with the dairy cluster, with three detected in downgradient wells: androsterone (WW-12, WW-15, and WW-17); androstenedienendione (WW-12); and testosterone (WW-11). The source of the androsterone and testosterone could be the dairy cluster, although androsterone was not detected in any of the dairy lagoons, manure piles, or application field samples. Androsterone is a metabolite of testosterone so could come

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from either humans or other animals, but in this case it was not detected in any of the other samples from the dairy cluster. Testosterone was detected in nine dairy lagoons and one manure sample and therefore its source could be from the dairy cluster or from a human source. The detection of androstenedienendione in the well is unexpected as it is a precursor to boldenone (a synthetic growth promoter) and is not intended for human use and is used mainly to treat horses and cattle.

The isotopic analysis indicates that the dominant nitrogen source for two wells is animal waste (WW-13 and WW-14) while the dominant source for WW-07 is fertilizer. For the other wells, it is not possible to determine the dominant source. For WW-08 and WW-15 the atmospheric contribution is strong.

In conclusion, all the downgradient residential water wells (with the exception of WW-10) associated with the Dairy Cluster have high nitrate levels. The data for total nitrogen and major ions indicate an increase in concentrations from the upgradient wells to the downgradient wells, with the likely sources being dairy lagoons, manure piles, and application fields.

The dominate nitrate source for the pharmaceuticals in WW-13 and WW-14 is animal waste and in WW-17 is predicted to be a mix of fertilizer and animal waste. Compounds were detected in these three wells that could be from sources associated with the dairy cluster, but could also be from human sources (tetracycline and oxytetracycline), with the exception of monesin.

It is possible ~~for that~~ the hormones ~~that the compounds found~~ in WW-12 or WW-17 came from sources associated with the dairy cluster. The isotopic evaluation predicted the source for WW-12 and WW-17 is probably a mix of fertilizer and animal waste. Again, a human source cannot be ruled out, as humans produce both testosterone and androsterone. For WW-11 and WW-15, it appears that the source is a mixture of fertilizer and atmospheric sources.

#### E.C. Residential Septic Systems

Instead of sampling septic systems directly, samples were collected from the influent stream of three small WWTP (Zillah, Mabton, and Toppenish) as surrogate samples for septic systems. This approach allows characterization of the typical compounds introduced with rural septage without having to sample the septic systems directly. This approach was used to determine whether the compounds found in the influent to the WWTP were similar or different to those found in downgradient water wells showing high nitrate concentrations in areas with significant numbers of septic systems.

Each of the five groups of compounds or analytical techniques is discussed for the septic systems. Because of the mixture of large numbers of different water sources to the flow entering the WWTP, age dating was not done on these samples. Four wells were specifically targeted to evaluate for linkage to septic systems (WW-19, WW-20, WW-21, and WW-22 – see **Figure 7**). However, while these four wells were targeted, all of the water wells were evaluated to determine whether septic systems could be the source of the nitrate found in any well in the study.

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## 1. Septic Systems: General Chemistry

### Septic Systems: Nitrate and other Forms of Nitrogen

The WWTP influents were not analyzed for nitrate because it was anticipated that there would be very little formation of nitrate from the organic nitrogen in the waste during its rapid transport to the treatment plant. This is the case because of the low oxygen environment of the sewer combined with the short residence time between the waste streams. The wells were evaluated for nitrate and the different forms of nitrogen. However, no analysis is possible because there are no upgradient wells to compare for comparison and no specific sources between the upgradient and downgradient wells.

### Septic Systems: Major Ions

Water quality parameters such as dissolved oxygen and conductivity were not collected from WWTPs because of concern that they could become easily contaminated. However, other studies (Pescod 1987) and a general knowledge of the nature of sewage indicates that sewage is very low in dissolved oxygen with high concentrations of dissolved solids compared with the groundwater or surface waters from which it is derived. Other general chemistry data were collected to characterize the WWTP influent as a surrogate for residential septic effluent. However, no pattern that might tie a specific well to a residential septic source is apparent when the major ion data from the treatment plants and water wells are compared.

As stated previously, the major ions for distinguishing waters are typically used to observe the evolution of water along a flow path. For the four wells identified as downgradient for the septic system analysis, upgradient wells that can be used to compare to downgradient wells were not available. Therefore, no specific analysis was conducted for the major ions.

### Septic Systems: Trace Elements

Four metals were found in the water wells and WWTPs (barium, iron, manganese, and zinc). However, each of these metals is naturally occurring elements and is not unique to septic systems. Any water sample would be expected to have detectable concentrations of each of these elements and the concentrations observed are similar for natural waters (Hem 1970).

### Septic Systems: Perchlorate

Perchlorate was analyzed to aid in the evaluation of the isotopic data. Perchlorate was not analyzed at the WWTP because it is not expected to persist in wastewater effluent as a result of bacterial activity and it is of value only when evaluating a particular water source, and not a mixture of sources such as is present in waste water. As stated earlier, perchlorate was used in this study as a potential indicator of the first flush of irrigation water moving through the caliche soils in the Lower Yakima Valley. The results for the perchlorate analysis are evaluated together with the isotopic data.

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## 19.2. Septic Systems: Microbiology

~~Similar to~~ As found with other water wells in the study, neither fecal coliform nor *E. coli* was detected in the four targeted water wells. The WWTPs were analyzed for fecal coliform and *E. coli*. (see Appendix A7). ~~As expected~~ Very high concentrations of both fecal coliform and *E. coli* were found in the influent to the WWTPs. Samples were also analyzed using MST to identify the source of the fecal contamination. Three of the samples were indicative of human sources, while one sample was indicative of both human and ruminant sources.

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## 20.3. Septic Systems: Organic Compounds

### Septic Systems: Pesticides

Samples were collected for analysis of pesticides in WWTPs by EPA's Manchester Laboratory. However, the laboratory reported that the WWTP sample matrix was too difficult to analyze because of significant interferences from the large number of organic compounds present in the waste. Therefore, the pesticide concentrations could not be quantified from the WWTP influent. No manure or application field samples are associated with these six sites.

Atrazine and bentazon were detected in WW-20 (Appendix A8). ~~Similar to the other wells with~~ atrazine and bentazon, the most likely source ~~of the pesticides~~ is past or current use of these compounds in agricultural production.

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### Septic Systems: Trace Organics

The trace organics were one class of compounds most likely to be associated with septic systems. The compounds sampled included those associated with human activities such as caffeine, fragrances, and disinfectants.

Thirty-seven trace organics were detected in the influent to the WWTPs (see Appendix A9). Nineteen of the trace organics were detected in all of the WWTP ~~samples~~. There were no detected trace organics in the four wells.

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### Septic Systems: Pharmaceuticals

Nine compounds were detected in the influent to the WWTPs for the wastewater pharmaceuticals, with six compounds detected in all three WWTPs influent ~~samples~~ (acetaminophen, cotinine, DEET, ibuprofen, naproxen, and tricolsan) (see Appendix A10). None of these compounds was detected in the four wells.

Table 19 shows the veterinary pharmaceuticals detected in the WWTP ~~influent~~ and the four wells targeted for evaluating septage. WW-22 contained no detected veterinary pharmaceuticals.

Table 19: Veterinary Pharmaceutical Detected in WWTP Samples

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Compound	WW-19	WW-20	WW-21	SP-01	SP-02	SP-03
Erythromycin	ND	ND	0.11	ND	ND	ND
Lincoymycin	ND	ND	0.371	ND	ND	ND
Monesin	0.194	ND	0.194	ND	ND	ND
Ractopamine	ND	ND	0.079	ND	ND	ND
Sulfachloropyridazine	ND	ND	0.334	ND	ND	ND
<b>Sulfamethazine</b>	ND	ND	0.053	ND	ND	0.086
<b>Sulfamethoxazole</b>	ND	ND	0.04	ND	0.106 (J)	0.662
Sulfathiazole	ND	ND	0.051	ND	ND	ND
<b>Tetracycline</b>	ND	242.6	ND	0.55 (J)	ND	ND
Tiamulin	ND	ND	0.05	ND	ND	ND
Virginiamycin	ND	ND	0.162	ND	ND	ND

Units = µg/L. Detection limit = 0.02 µg/L.

J values mean the compound was positively identified, but the associated numerical value is an estimate.

ND = Not detected.

Three compounds were detected in the water wells and at least one WWTP influent (sulfamethazine, sulfamethoxazole, and tetracycline). Eight compounds were detected in the water wells, but not in the WWTPs. WW-21 had the highest number of compounds detected (10) of any water well. Water well WW-21 is surrounded by possible septic sources; it is also downgradient from several hop yards and although at a greater distance, also downgradient from several large dairies. These factors make it challenging to identify the potential source of the veterinary pharmaceuticals at WW-21.

Many of the compounds detected in the WW-21 were not found in the WWTPs. Many of these compounds are used by humans (for example, tetracycline, lincomycin, and the sulfonamides), and it is possible that they are excreted and possibly end up in septic systems. In this case, it would be anticipated that they would be detected in WWTPs. One possible explanation is that the WWTP matrix is much more complex, with an increased possibility of interferences than the cleaner water wells. When large numbers of organic molecules are present, it makes it difficult to detect these compounds in the WWTP influent.

#### Septic Systems: Hormones

EPA's Ada laboratory analyzed all four water wells and the WWTP influents for five hormones.. The laboratory did not detect any of the five hormones in the water wells, but did detect three of the hormones in WWTP samples: 17-β-estradiol, estriol, and estrone (**see Appendix A12**).

The UNL analyzed samples for the four water wells and the WWTPs for 18 hormones, including the same five hormones as Ada (**see Appendix A13**). No hormones were detected in WW-19 and WW-21. Androsterone was detected in WW-20. Eight compounds were detected in WW-22. **Table 20** shows the concentrations of the compounds detected in these wells and their corresponding concentrations in the WWTPs.

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**Table 20: Results for Hormones from UNL**

Compound <sup>(*)</sup>	WW-20	WW-22	SP-01	SP-02	SP-03
17-β-estradiol	ND	<b>0.006</b>	<b>0.012</b>	<b>0.035</b>	<b>0.034</b>
α-estradiol	ND	<b>0.005</b>	<b>0.263</b>	ND	ND
Androsterone	<b>0.004(J)</b>	ND	<b>5.049(J)</b>	<b>2.137(J)</b>	<b>3.187(J)</b>
Androstenedienendione	ND	<b>0.005</b>	<b>0.255(J)</b>	<b>0.614(J)</b>	<b>14.1 (J)</b>
β-Zearalanol	ND	<b>0.003</b>	ND	ND	ND
Estrone	ND	<b>0.004</b>	ND	ND	ND
Testosterone	ND	<b>0.01</b>	<b>0.053</b>	<b>0.059</b>	<b>0.045</b>
Keto Testosterone	ND	<b>0.005</b>	<b>0.1</b>	<b>0.043</b>	ND
Epitestosterone	ND	<b>0.004</b>	ND	<b>0.06</b>	ND

\*Both Ada and UNL analyzed for 17-β-estradiol, α-estradiol, and estrone. UNL detected 17-β-estradiol, α-estradiol, and estrone in WW-22. Only Ada detected these two compounds for the WWTP (SP01-SP03). UNL analyzed for the other compounds.

Units = µg/L. Detection limit = 0.02 µg/L. J values mean the compound was positively identified, but the associated numerical value is an estimate. ND = Not detected.

WW-20 is not located in close proximity to a dairy, so the potential source of androsterone in these samples could be a septic system or another source.

WW-22 had detections of multiple compounds. Several of the compounds detected in WW-22 were also detected in the WWTPs (17-β-estradiol, α-estradiol, testosterone, keto-testosterone, and epitestosterone). These compounds would be expected to be detected in WWTP given they are natural sex hormones or are produced in mammals. WW-22 is not in close proximity to a dairy, which would reduce the chance that the source of these compounds is a dairy, although the agricultural land use upgradient of the well could be using dairy manure as fertilizer. It is possible that the compounds detected in WW-22 are coming from septic systems that are in the vicinity of this well.

#### 21-4 Septic Systems: Isotopic Analysis

Isotopic analysis was completed for all water well samples. Table 20 provides the results for these four wells.

Location	Nitrate -N (mg/L)	δ <sup>15</sup> N- NO3 (‰)	Dominant Source	δ <sup>18</sup> O-NO3 (‰)	Overall Assessment
WW-19	36.4	8.7	Fertilizer & Animal Waste	15.4	Fertilizer & Animal Waste
WW-20	15	6.3	Fertilizer & Animal Waste	52.9	Fertilizer & Animal Waste & Atmospheric
WW-21	36.5	7.7	Fertilizer & Animal Waste	12.2	Fertilizer & Animal Waste
WW-22	16.6	10	Animal Waste	11.0	Animal Waste

[ PAGE \\* MERGEFORMAT ]

$\delta^{15}\text{N-NO}_3$ . Values less than 2.0 = dominated by fertilizer; values between 2.0 to 8.4= undetermined mixture of fertilizer and/or animal waste; values greater than 8.4 = dominated by animal.  
 $\delta^{18}\text{O-NO}_3$ . Values greater than 20 considered strong atmospheric contribution.

The dominant source of nitrate in WW-22 appears to be animal waste. For the other water wells, the potential sources are likely to be a combination of fertilizer and/or animal waste for WW-19 and WW-21 and a combination of fertilizer and/or animal waste with a strong atmospheric contribution for WW-20. The probable sources of nitrate for these water wells match the variety of land uses surrounding these highly scattered water wells.

#### 22.5 Septic Systems: Age Dating

Table 21 provides the age dating results for all four water wells.

**Table 21: Septic Systems – Summary of Age Dating Analyses for WW-19 to WW-22 (Years)**

Location	Sample Age	Duplicate Age	Average
WW-19	44.3	34.3	39.3
WW-20	14.3	14.3	14.3
WW-21	31.3	28.8	30.1
WW-22	29.3	29.3	29.3

There is a wide scatter of ages in the water wells with age measurements ranging from 14.3 years to 44.3 years with no discernible spatial pattern.

#### 22.6 Septic Systems – Summary of Results for Residential Water Wells

Table 22 provides a summary of the groups of compounds (general chemistry and organic compounds) and analytical techniques (isotopic analyses) that provide information useful to address the question of the likely sources of the nitrate for the four residential water wells associated with the septic systems. No conclusions using the microbial data is possible given the downgradient wells did not exhibit any microbial contamination. In addition, the age dating data do not provide any specific evidence to connect specific sources to high nitrate levels.

**Table 22: Septic Systems – Summary of Results for Residential Water Wells**

General Water Chemistry	Organic Compounds Detected	Organic Compounds Detected in WWTPs	Isotopic Analyses
<b>WW-19</b>			
Nitrate level = 38.2 mg/L	Monesin	Monesin not detected in WWTPs	Fertilizer & Animal Waste
<b>WW-20</b>			

Nitrate level = 15 mg/L	Atrazine and bentazon	Atrazine and Bentazon not analyzed	Animal waste & Fertilizer & Atmospheric
	Tetracycline	Tetracycline (SP-01)	
	Androsteronene	Androsterone (All WWTPs)	
WW-21			
Nitrate level = 38 mg/L	Erythromycin, lincomycin, monesin, ractopamine, sulfamethazine, sulfamethoxazole, sulfathiazole, tiamulin, and virginiamycin	Sulfamethazine (SP-03)  Sulfamethoxazole (SP-02 and SP-03)  Others not detected in WWTP influent	Fertilizer & Animal Waste
WW-22			
Nitrate level = 16.4 mg/L	11-Keto testosterone	11-keto Testosterone (SP-01; SP-02)	Animal waste
	17-β-estradiol α-estradiol, Androstendienendione	17-β-estradiol (All WWTPs) α-estradiol (SP-01) Androstendienendione (All WWTPs)	
	β-zearalanol Estrone Testosterone Epitesterone	β-zearalanol (No detects) Estrone (No detects) Testosterone (All WWTPs) Epitesterone (SP-02)	

The main information anticipated to provide a connection with septic systems was from the organic compounds, microbiological data, and possibly the isotopic data. The major ions, minor and trace inorganic elements, and forms of nitrogen analyses are not useful because of the highly variable results from these spatially distributed wells of unknown construction. As discussed above, the trace inorganic elements are ubiquitous in the environment and highly variable in concentration.

In addition, these wells were sampled in isolation – that is, without a pairing with an upgradient well with a specific source separating them. For this reason, no chemical or temporal evolution along a flow path can be demonstrated from these data. Age dating results are similarly highly variable, likely for the same reasons of spatial distribution and unknown well construction.

The pesticides atrazine and bentazon were detected in WW-20. Trace organics were the class of compounds anticipated to provide data to possible link to septic systems. There were no detections of any trace organics for the water wells targeted for the septic systems. Five trace organic compounds were found for all the water wells in the study: bis-(2-ethylhexyl) phthalate

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(DEHP); 5-methyl-1h-benzotrizole; naphthelene; tetrachloroethylene, and phenol. DEHP was the only trace organic detected in the three WWTPs. DEHP was the only trace organic detected in any of the residential water wells. The other trace organics were detected in dairy supply wells.

For pharmaceuticals, WW-21 had the highest number of compounds (10) detected of any wells. WW-21 is surrounded by possible septic sources; it is also downgradient from several hop yards and also at a greater distance downgradient from a dairy. The isotopic data indicate that the source of nitrate is a probably a combination of fertilizer and animal waste.

WW-22 had detections for eight compounds for hormones. WW-22 is not in close proximity to a dairy, which would reduce the chance that the source is a dairy, although the agricultural land upgradient of the well could be using dairy manure as fertilizer. It is also possible that the detections for WW-22 are from a septic system, although the detection of androstenedienendione and  $\beta$ -Zearalanol was not expected given they are not approved for human or dairy cow use. However, androstenedienendione was detected in the WWTP, indicating there is a source within the area for this compound.

In conclusion, all of the residential wells had high levels of nitrate. Other evidence to link septic systems are the pharmaceutical, hormone, and isotopic data. Compounds were detected in several wells that are associated with humans.

#### G.D. Irrigated Cropland

Another likely source of nitrates is irrigated croplands. The likely inputs examined in this study are inorganic fertilizer and manure applied to the land from dairies. This study looked at three crops: mint, hops, and corn. Soil samples were collected from six fields that were located upgradient from six residential drinking water wells. Corn and hop fields typically receive both manure and synthetic fertilizer inputs at different times during the year. Mint fields typically receive only synthetic fertilizer. Each soil sample and associated water well sample is shown in **Figure 7 and Table 23**.

**Commented [LE69]:** In the description of the 3 study phases, the potential sources are described in a different order—dairies, irrigated cropland, then septic systems and other. Why not use the same order when describing results?

**Table 23: Irrigated Cropland – Soil Samples and Associated Water Wells**

Soil Sample	Associated Water Well	Crop
SO-11	WW-23	Mint
SO-12	WW-24	Mint
SO-13	WW-25	Corn
SO-14	WW-28	Corn
SO-15	WW-26	Hops
SO-16	WW-27	Hops

The soil samples were analyzed for: nitrogen species, pesticides, pharmaceuticals, and hormones. They were not analyzed for general chemistry, microbiology, trace organics, isotopic analysis, or age dating. ~~It was concluded that~~ the compound classes evaluated would provide the best information to link irrigated cropland with high nitrate levels in nearby water wells. Some of the analyses are not usually performed on soil samples (perchlorate, major ions, isotopic analysis, and age dating), while others were not completed because of resource constraints (trace organics).

## 1. Irrigated Cropland: General Chemistry

### Irrigated Cropland: Nitrate and Other Forms of Nitrogen

The water wells associated with the irrigated cropland had high levels of nitrate and were all above the nitrate MCL of 10 mg/L. Soil samples were analyzed for several forms of nitrogen, including extractable nitrate, extractable ammonium, and total nitrogen by combustion. The values measured in the soil samples for extractable nitrate (nitrate-N) and extractable ammonium (ammonia-N) can be compared with limits recommended by the Washington Department of Agriculture for fields under National Pollutant Discharge Elimination System permits. The recommendations are that the concentration of nitrate-N in the post-harvest soil should be no more than 45 milligrams per kilogram (mg/kg) or parts per million. **Table 24** shows the measured values for these forms of nitrogen in these crop soils.

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these sample locations were selected

**Table 24: Irrigated Cropland – Concentrations of Nitrogen Forms in Soil Samples**

Soil Sample/Crop	Nitrate-N (ppm)	Ammonium-N (ppm)	Total N by Combustion (ppm)
SO-11 – Mint	245	210	3330
SO-12 – Mint	191	8.2	2350
SO-13 – Corn	24.3	7.5	1100
SO-14 – Corn	6.3	12	1180
SO-15 – Hops	83.5	21	2210
SO-16 – Hops	26.5	7.7	3000

The cropland soil samples were collected at the beginning of the growing season. As such, they are not directly comparable to post-harvest soil monitoring guidelines. However, values of nitrate-N measured in the soils during the study were five times the guidelines. These levels, if not reduced by either uptake by growing plants, volatilization, or denitrification, would ultimately move out of the root zone where they could become a source of groundwater contamination.

## 24.2 Irrigated Cropland: Organic Compounds

### Irrigated Cropland: Pesticides

Fifteen pesticides were detected in one or more of the six soil samples targeted for crops (see **Appendix A8**). Several pesticides were detected in more than two soil samples (2,4-D was detected in four soil samples; 4-nitrophenol in six soil samples; and pentachlorophenol in five soil samples).

Of the 15 pesticides detected in the six soil samples, only two were detected in the six water wells targeted for impact from crops: bentazon (WW-23 and W-24) and atrazine (WW-24 and WW-26). **Table 25** indicates that bentazon was detected in soil samples related to water wells at two of the three associated sites: SO-11/WW-23 and SO-12/WW-24. Atrazine was not detected in any of the soil samples associated with the water wells that had detections of atrazine.

**Table 25: Irrigated Cropland – Concentrations of Pesticides in Soil Samples and Associated Water Wells (soil sample values are reported first and then water well associated with the soil samples are reported in each cell)**

Compound	Mint SO-11 WW-23	Mint SO-12 WW-24	Corn SO-13 WW-25	Corn SO-14 WW-28	Hops SO-15 WW-26	Hops SO-16 WW-27
Atrazine	ND ND	ND <b>0.017 (J)</b>	<b>1.6(J)</b> ND	<b>0.7(J)</b> ND	ND <b>0.025(J)</b>	ND ND
Bentazon	<b>38</b> <b>0.028 (J)</b>	<b>2 (J)</b> <b>0.033(J)</b>	ND ND	ND ND	ND ND	ND ND

Units for water wells = µg/L. Units for soil samples = Micrograms per kilogram (µg/kg).

ND = Not detected. Detection limit for soil and water well samples vary depending on the analysis.

“J” values mean the compound was positively identified, but the associated numerical value is an estimate.

Given the widespread use of both atrazine and bentazon, it is likely that the detections in the water wells are ~~seeing the result of~~ past and current use of these pesticides.

### Irrigated Cropland: Pharmaceuticals

Veterinary pharmaceuticals were detected in one well (WW-26). Nine compounds were detected. No veterinary pharmaceuticals were detected in the other wells. Monesin was the only compound detected in both a water well (WW-26 at 0.319 µg/L) and its associated soil sample (SO-15 at 4.5 micrograms per kilogram [µg/kg]). Veterinary pharmaceuticals were detected in five of the soil samples, but these compounds were not detected in the associated water well samples (SO-13 had no detections).

In addition to monesin, erythromycin, lincomycin, ractopamine, sulfamethazine, sulfamethoxazole, sulfathiazole, tiamulin, and virginiamycin were detected in WW-26. None of these compounds was detected in the soil samples.

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WW-26 is surrounded by hop yards that receive manure applications. There is also a dairy generally upgradient from the well, although it is at some distance. It is possible that the compounds detected in WW-26 came from the application of manure. This would be especially true for monesin. However, it is also possible that at least some compounds came from septic fields, given that several of the compounds are used by humans.

#### Irrigated Cropland: Hormones

Water well WW-27 was the only water well with any compounds detected. There were three compounds detected in WW-27 (androstenedione, 17- $\beta$ -trenbolone, and testosterone). These compounds were not detected in the soil sample (SO-16) associated with WW-27.

Five hormones were detected in soil samples (see Appendix A13).

- \* SO-11: Androstenedione and progesterone
- \* SO-12: Androstenedione, 17- $\alpha$ -estradiol, and progesterone
- \* SO-13: Melengesterol acetate
- \* SO-15: 4-androstenedione and androstenedione
- \* SO-16: 4-androstenedione and progesterone

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All of the hormones in the soil samples (except for melegensterol acetate) are naturally produced by animals and can be expected to be found in septic systems, manure piles, and dairy lagoons.

#### 2.3.3. Irrigated Cropland: Isotopic Analyses

An isotopic analysis was completed for six wells associated with the irrigated croplands. One mint field (WW-24) and one corn field (WW-25) showed nitrate originating predominantly from a fertilizer source. Fertilizer and atmospheric deposition were the dominant sources for WW-23. Water well WW-26 had a mixed animal waste and fertilizer source, while water wells WW-27 and WW-28 had a dominant source of fertilizer, animal waste, and atmospheric (Table 26).

**Table 26: Irrigated Cropland – Summary of Isotopic Analysis for Water Wells**

Location	Nitrate-N (mg/L)	$\delta^{15}\text{N}$ - NO3 (‰)	Dominant Source	$\delta^{18}\text{O}$ - NO3 (‰)	Overall Assessment
WW-23 (Mint)	17.3	2.2	Fertilizer & Animal Waste	18.0	Fertilizer & Animal Waste
WW-24 (Mint)	14	-0.3	Fertilizer	12	Fertilizer
WW-25 (Com)	32.9	2.4	Fertilizer & Animal Waste	15	Fertilizer & Animal Waste
WW-26 (Hops)	15.1	7.5	Fertilizer & Animal Waste	6.3	Fertilizer & Animal Waste

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WW-27 (Hops)	19.9	8.8	Animal Waste	17	Animal Waste
WW-28 (Corn)	69.6	5.5	Fertilizer & Animal Waste	44	Fertilizer & Atmospheric & Animal Waste

$\delta^{15}\text{N-NO}_3$ . Values less than 2.0 = dominated by fertilizer; values between 2.0 to 8.4 = undetermined mixture of fertilizer and/or animal waste; values greater than 8.4 = dominated by animal.  
 $\delta^{18}\text{O-NO}_3$ . Values greater than 20 considered strong atmospheric contribution.

The dominant source for WW-24 is fertilizer while the dominant source for WW-27 is animal waste. For the other water wells, the potential sources are likely to be a combination of fertilizer and animal waste for WW-23, WW-25, and WW-26 and a combination of fertilizer and animal waste with a strong atmospheric contribution for WW-28.

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#### 26.4. Irrigated Cropland: Age Dating

The age dating date for the six wells associated with the irrigated crops is presented in Table 27.

**Table 27: Irrigated Cropland – Summary of Age Dating Analyses for Water Wells (Years)**

Location	Sample Age	Duplicate Age	Average
WW-23: Mint Field	Over Value	Over Value	NA
WW-24: Mint Field	14.8	15.8	15.3
WW-25: Corn Field	10.3	9.8	10.1
WW-26: Hops Field	12.8	11.8	12.3
WW-27: Hops Field	Over Value	14.3	14.3
WW-28: Corn Field	Over Value	Overvalue	NA

Over Value: These samples contained more  $\text{SF}_6$  than can be explained by equilibrium with modern air.

**Commented [LE72]:** This is the explanation I was looking for earlier, even though I'm not sure what it means

While downgradient wells were paired with soil samples from the crop fields, they were not paired with wells upgradient of crop fields. For this reason, no pattern emerges from review of the age dating data beyond the observation that the values are younger than for any other group of samples in the study. This finding could be due to the very young age of the ~~because~~ irrigation water was used on the crop fields and the very young age of that water.

#### 27.5. Irrigated Cropland – Summary of Results for Residential Wells

Table 28 provides a summary of the groups of compounds (general chemistry and organic compounds) and analytical techniques (isotopic analyses) that provide information useful to address the question of the likely sources of the nitrate for the four residential water wells associated with the septic systems. No conclusions using the microbial data is possible given the downgradient wells did not exhibit any microbial contamination. In addition, the age dating data do not provide any specific evidence to connect specific sources to high nitrate levels.



**Table 28: Irrigated Cropland – Summary of Results for Residential Water Wells**

<b>WW-23 and SO-11 (mint)</b>			
<b>General Chemistry</b>	<b>Organic Compounds in Water Wells</b>	<b>Organic Compounds Also in Associated Soil Samples</b>	<b>Isotopic Analyses</b>
Nitrate level = 16.0 mg/L	Bentazon	Bentazon	Fertilizer & Animal Waste
<b>WW-24 and SO-12 (Mint)</b>			
Nitrate level = 13.8 mg/L	Atrazine Bentazon	Bentazon	Fertilizer
<b>WW-25 and SO-13 (Corn)</b>			
Nitrate level = 33.4 mg/L	No detects	Nothing to compare	Fertilizer & Animal Waste
<b>WW-26 and SO-15 (Hops)</b>			
Nitrate level = 15.3	Atrazine, erythromycin, lincomycin, monesin, ractopamine, sulfamethazine, sulfathiazole, itamulin, and virginiamycin	Monesin only compound also detected in soil sample	Fertilizer & Animal Waste
<b>WW-27 and SO-16 (Hops)</b>			
Nitrate level = 19.8 mg/L	Androstadienedione 17- $\beta$ -Trenbolone Testosterone	These compounds were not detected in soil samples	Animal Waste
<b>WW-28 and SO-14 (Corn)</b>			
Nitrate level = 71.2 mg/L	No detects	Nothing to compare	Fertilizer & Atmospheric & Animal Waste

Bentazon and atrazine were the only pesticides detected in the water wells associated with the six soil samples. Bentazon was detected in the soil samples associated with the water wells at two sites: SO-11/WW-23 and SO-12/WW-24. This is significant and indicates that the bentazon applied to the crop fields is being associated with concentrations in nearby wells. Atrazine was not detected in any of the soil samples associated with the water wells with detections of atrazine.

WW-26 was the only well with detected concentrations for the pharmaceuticals (eight compounds detected). None of these compounds was detected in the associated soil sample (SO15). WW-26 is surrounded by hop yards that receive manure applications. There is also a dairy generally upgradient from the well, although it is at some distance. It is possible that the compounds detected in WW-26 came from the application of manure. However, it is also possible that they came from septic fields, given these compounds are used by humans.

Three hormones were detected in one well (WW-27). The three hormones were not detected in the associated soil sample (SO-16).

In conclusion, several compounds were detected in the water wells (WW-23, WW-24, WW-26, and WW-27). However, with the exception of bentazon, none of these compounds was detected in the associated soil samples.

#### **Water Wells WW-18 and WW-30**

Two other residential water wells were evaluated: WW-18 and WW-30. WW-18 was sampled because the owner was aware of the study and volunteered his property for sampling. WW-30 was sampled because it was located in an area not otherwise sampled and was high in nitrate.

WW-18 was analyzed for all the compounds and for isotopic and age dating. WW-30 was not evaluated for hormones, pharmaceuticals, isotopic, or age dating as the site was added later in the study. The results for the two wells are included in Table 29.

**Table 29: Results for WW-18 and WW-30**

Compounds	WW-18	WW-30
Nitrate	72.2 ug/L	23.4 ug/L
Microbiology	No detects	No detects
Organic Compounds	Atrazine, tetracycline, and testosterone	Atrazine, bentazon, and phenol
Isotopic Analysis	Fertilizer & Animal Waste	Not conducted
Age Dating	28.1 years	Not conducted

While the major ions and different nitrogen forms were measured for both of these samples, the results are not included because they are used to observe the evolution of water along a flow path. There were no upgradient wells that can be used to compare and no specific source with which to compare for WW-18 and WW-30. Therefore, no specific analysis was conducted for the major ions and total nitrogen.

Neither fecal coliform nor *E. Coli* were detected in the WW-18 or WW-30. Atrazine, tetracycline, and testosterone were detected in WW-18. Atrazine, bentazon, and phenol were detected in WW-30. Phenol was abundant in the dairy lagoons sampled and can also be found in household wastewater. The source of the phenol in the water well could be septic systems. WW-30 is not located in the vicinity of a dairy. WW-30 was not analyzed for wastewater pharmaceuticals based on its late addition at the end of the study.

## X. STUDY LIMITATIONS AND UNCERTAINTIES

There were several constraints in the study that are important to highlight. Primary sources of uncertainty stem from inadequate or absent information on well construction, the lack of standard analytical methods for all parameters, and the absence of specific knowledge of dairy operations. Combined with the complex matrices of some of the samples, these factors limit the certainty with which connections can be made between specific sources and downgradient sampling locations. -

**Commented [LE73]:** I think it would help to introduce them by type before plunging into the more detailed explanations. I'll give it a shot, but you can probably do better. I got them a little out of order from the descriptions that follow, though—because the first 3 were lack of info, they went together.

**Commented [LE74]:** Or something like that. There is probably a better way to phrase this last sentence.

First and perhaps most importantly, the well depths, screened intervals, and construction details of the wells sampled were generally unknown. An attempt was made to locate the wells logs, but it was not possible to match the well log information with the specific sampling sites in the study for the majority of the wells. For this reason, it is generally unknown whether the water chemistry in water wells represents shallow groundwater, deeper groundwater, or more than one zone being mixed.

Several of the wells sampled showed both low dissolved oxygen and high nitrate. This result is an indication of mixing of waters of different chemistries since nitrate is rapidly depleted in waters after oxygen is consumed by bacterial activity. More specific information on each well would enable confirmation of the hydraulic connection with the upgradient well, sources, and downgradient wells using the water chemistry for each well because the wells are potentially tapping different flow paths or simply mixing waters of unrelated chemistries.

Second, many of the analytical methods utilized in the study were EPA standard methods (nitrate and other nitrogen forms; trace elements; major ions, and pesticides). However, some of the analytical methods used are not EPA or equivalent standard methods, but are used primarily for research purposes (pharmaceutical and hormones). The research methods use complex instrumentation technologies and are capable of detecting trace levels of the target compounds.

**Commented [LE75]:** This doesn't sound like a limitation or uncertainty the way it is worded. Is it that it might not be easily replicated or what?

As can be expected, however, variability with quality control results was observed because of matrix effects. Modifications to the procedures were implemented by the laboratories as needed to improve chromatography and detection. In addition, the combination of very low detection limits and complex matrices may yield false positives or the inability to see low concentrations despite the appropriate use of blanks, matrix spikes, surrogates and duplicates. For these reasons, despite the best efforts of the laboratories selected to conduct this work, the data generated may not be reproducible by a third party, one of the criteria required by EPA's Information Quality Guidelines.

**Commented [LE76]:** Is this statement intended to apply to all the samples or some specific analyses or sample types?

Finally, there was limited information regarding the dairy operations. EPA had requested information on specific aspects of the dairy operations to develop a better understanding of their day-to-day operations, but that request was denied. The lack of specific information on dairy operations creates some uncertainty regarding the relationship between specific compounds detected in water wells and the dairies (for example, whether the dairy used specific pharmaceuticals for treatment of animals).

**Commented [LE77]:** Could this one be moved up to join the other "lack of info" list?

## XI. CONCLUSIONS

This report presents the results for sampling conducted from February 2010 to April 2010 by the U.S. Environmental Protection Agency (EPA) in the Lower Yakima Valley in Central Washington State. The primary purpose of this study was to investigate the contribution of various sources from nearby land uses to the high nitrate levels in drinking water wells. The study looked at three likely sources of nitrate: dairies; irrigated cropland; and residential septic systems and private residential drinking water wells for a variety of chemicals to determine if chemicals other than nitrate can be used to link the nitrate contamination in groundwater to specific sources. The analyses included chemicals that are expected to be associated with one or more of the likely sources, such as pharmaceuticals (both veterinary and human medications), personal care products, steroids and hormones, pesticides and herbicides, as well as other indicators of water quality.

The investigation also used microbial analysis to determine whether the drinking water wells were contaminated with fecal contamination. If the water wells were found to have fecal contamination, then Microbial Source Tracking (MST) was performed to identify the source (i.e., human or ruminant) of the fecal contamination. In addition, EPA performed isotopic analysis for the water wells to determine the general source, or combination of sources, of nitrates in the water wells. Finally, an age dating analysis was completed for the water wells to determine the time since infiltration of water into the water wells.

In general, the microbial and age dating data did not provide information that could be used to help identify specific sources that can contribute to the high nitrate levels detected in residential drinking water wells.

The ~~best strongest~~ evidence ~~to link~~ linking specific sources to the high nitrate values was the total nitrogen and major ion data associated with the dairies. This data showed that the dairies were likely contributing to the increased trend in total nitrogen and major ion concentrations between the upgradient wells ~~to and~~ the downgradient wells. The specific contribution from the dairies is difficult to ~~determine~~ because of the lack of information on water wells.

Commented [LE78]: Do you mean "quantify"?

Many of the organic compounds (e.g., pharmaceuticals and hormones) analyzed were detected in the dairy lagoons, manure piles, and application fields as well as the WWTP ~~influent used as septic surrogates~~. In general, the majority of these compounds were not detected in the residential water wells with several notable exceptions (WW-21 with 10 compounds detected and WW-22 with nine compounds detected).

The ~~presence of these compounds~~ pharmaceuticals and hormones in ~~these sources at dairies and in WWTP influent~~ indicates they are used at the dairies or by humans (WWTPs) but were not transported or were not transported at detectable levels to the downgradient wells during the study period. This could be the case because organic compounds are typically less mobile in water than inorganic compounds. Organic compounds tend to adsorb to organic carbon in the aquifer material and may be degraded by bacteria and either disappear entirely or may be greatly reduced

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in concentrations. Even if not broken down, most organic compounds will move much slower than nitrate because they tend to adsorb to other organic matter in the aquifer. As a result, in general, they are unlikely to be transported as far or as fast as the nitrate.

The isotopic data provide evidence that the dominant source of nitrate is animal waste for four residential wells (WW-05, WW-13, WW-14, and WW-22) and the dominant source of nitrate is synthetic fertilizer for one residential water wells (WW-24). The specific ~~single~~ source of nitrate for the remainder of the residential water wells was not determined, but attributed to a combination of synthetic fertilizer and/or animal waste. Four residential water wells were determined to have a strong atmospheric contribution (WW-03, WW-15, WW-20, and WW-28).

Commented [LE79]: If that's what you mean

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**APPENDIX A: DATA SUMMARY TABLES (included in a separate excel  
spreadsheet)**

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## APPENDIX B: DETAILS ON THE ISOTOPIC ANALYTICAL RESULTS OF THE STUDY

Samples from all the water well, dairy lagoon, and WWTP influent were submitted to the University of Nebraska, Lincoln Laboratory for isotopic analysis. The results of the isotopic analyses are presented in **Appendix A14**. The isotopic analysis is used to assist to identify the general source, or combination of sources, or dominant processes that have contributed nitrates to the drinking water wells evaluated in this study. Most of the literature on isotopic fractionation, particularly the part attributing specific samples to specific sources, makes clear that the science is still evolving and that this tool is most appropriately used to supplement other methods used to investigate the source of nitrates (Kendall and others 2007).

The sources evaluated for the isotopic analysis include nitrate formed locally in soil derived from breakdown of plant material; synthetic fertilizers; animal waste, including humans; and accumulation from atmospheric deposition from precipitation and dry deposition. It is important to note that the animal waste source does not differentiate between humans and other animal wastes.

The location of the Lower Yakima Valley at the base of the Cascade Mountain Range and in the rain shadow of those mountains has implications that lead to predictable variations in isotopic. This location leads the U.S. Environmental Protection Agency (EPA) to expect deviation from some of the patterns seen in other collections of data areas. For example, because of the very low rainfall in this area, it is anticipated that atmospherically deposited nitrate would accumulate in shallow soils in the caliche layer. This A caliche layer is characteristic of desert regions and forms when carbonate minerals accumulate in the shallow subsurface because insufficient rainfall occurs to wash them into the deeper groundwater. Other minerals may accumulate along with the carbonates in areas of very low rainfall. These include gypsum and, if the area is sufficiently dry, nitrates and perchlorates, a highly oxidized form of chlorine.

**Commented [LE80]:** Not unique—there are plenty of other rain shadows

EPA tested for perchlorates in addition to nitrate to explore the possibility of natural buildup of nitrate in valley soils from atmospheric deposition. The reason for testing for perchlorate was to identify areas where recent addition of irrigation water could be acting to flush a reservoir of soluble compounds such as nitrate and perchlorate out of these desert soils.

EPA followed a two-step process to link a dominant process or a specific potential nitrogen source to each water well, dairy lagoon, and wastewater treatment plants (WWTPs) sampling location. First, the concentrations of nitrate in each sample were compared with values typical of systems where nitrate is locally derived from natural plant breakdown in soil. The purpose was to determine whether locally derived soil nitrate was a likely source of the nitrate.

Naturally occurring nitrate concentrations are generally expected to range from 0.3 to possibly 1.1 milligrams per liter (mg/L) (Nolan and Hitt 2003). Nitrate concentrations observed in all downgradient wells (as opposed to “supply” or upgradient well”) were in excess of the value that could be expected from natural soil nitrate formation (**See Table A4 in Appendix A**). The high

concentrations of nitrate in all of the downgradient well samples indicate that locally derived soil nitrate ~~from~~ the breakdown of local growing plants is not a likely process to explain the data in this study.

The second step was to look for plausible sources and evaluate each sample based on those ~~potential~~ sources. For animal waste, data from the dairy lagoons were used to evaluate the ~~potential values results~~ that would indicate the dominant source is from animal waste (Table B-1).

Three dairy lagoon samples were collected from each dairy. One sample was collected at the freshest inlet end of the dairy lagoon system. For example, LG-01 was sampled at the “freshest” or influent end of the dairy lagoon system. LG-02 and LG-03 were collected at the discharge end of the system just before it was pumped onto the spray-irrigation fields. Samples LG-01, LG-04, LG-07, LG-10 and LG-13 were intended to show the youngest and least volatilized waste.

The expected trend would be for waste to increase in  $\delta^{15}\text{N-NH}_4$  as the light ammonia is preferentially volatilized and the residual in the dairy lagoon system is enriched in the heavier form. This trend is ~~seen evident in the data~~ with the first of each group of three dairy lagoon samples being a smaller number (lighter) and the last two being heavier. Lagoon samples LG-10 to LG-12 differ from the other ~~later~~ dairy lagoons because they seem to lack any sign of volatilization or maturation during passage through the system. This pattern may be related to the location where EPA sampling teams collected the sample or the rate at which waste passes through this system.

**Commented [LE81]:** What would make a lagoon “later” as opposed to just “other”? Or is this about the samples? Then it should say “differs from later samples”

The last two samples were collocated in each of the dairy lagoons, with the exception of LG-15, which is from a different dairy lagoon at the most distant portion of the system. The co-located samples (LG-02 and LG-03; LG-05 and LG-06; LG-08 and LG-09; and LG-11 and LG-12) all show similar isotopic values. Dairy lagoon sample LG-15 shows a much “heavier” signature for  $^{15}\text{N}$  for the remaining ammonia, as is expected from its location at the distal end of the dairy lagoon system on the Bosma Dairy.

The average  $\delta^{15}\text{N-NH}_4$  of the five influent streams to the dairy lagoons in this study (LG-01, LG-04, LG-07, LG-10, and LG-13) was 5.0‰. The average of the  $\delta^{15}\text{N-NH}_4$  in the 10 dairy lagoon ~~values samples in the older material immediately prior to before~~ land application (LG-02, LG-03, LG-05, LG-06, LG-08, LG-09, LG-11, LG-12, LG-14, and LG-15) was 8.4‰.

**Table B-1: Isotopic Analysis - Summary Results for Lagoons**

Location	Position in System	Ammonia (mg/L)	$\delta^{15}\text{N-NH}_4$ (‰)	Assessment
LG-01: Haak	Influent	907	3.4	Fresh Animal Waste
LG-02:Haak	Discharge	923	10.1	Volatilized animal waste
LG-03: Haak	Discharge	896	9.9	Volatilized animal waste
LG-04: DeRuyter	Influent	899	6.7	Volatilization Animal Waste
LG-05:DeRuyter	Discharge	1151	10.6	Volatilization Animal Waste
LG-06: DeRuyter	Discharge	1293	10.3	Volatilization Animal Waste
LG-07:D&A	Influent	869	5.4	Fresh Animal Waste
LG-08: D&A	Discharge	696	10.3	Volatilization Animal Waste
LG-09: D&A	Discharge	658	10.1	Volatilization Animal Waste
LG-10: Cow Palace	Influent	NM	NM	NM
LG-11:Cow Palace	Discharge	274	3.1	Fresh Animal Waste
LG-12: Cow Palace	Discharge	222	2.0	Fresh Animal Waste
LG-13:Bosma	Influent	469	4.4	Fresh Animal Waste
LG-14:Bosma	Discharge	600	3.3	Fresh Animal Waste
LG-15:Bosma	Discharge	658	13.9	Volatilization Animal Waste

Based on this data, EPA decided that a  $\delta^{15}\text{N-NO}_3$  value of 8.4‰ in water wells was a reasonable value to indicate that the source of nitrate was dominated by animal waste. This does not mean that all animal waste has  $\delta^{15}\text{N-NO}_3$  values above 8.4‰. EPA believes it does indicate that values above this are in the range in which nitrate isotopes can be used to identify animal waste derived from nitrate with reasonable confidence. The value of 8.4‰ for  $\delta^{15}\text{N-NO}_3$  is relatively consistent with information from the literature indicating a general range of  $\delta^{15}\text{N-NO}_3$  values between 10‰ and 20‰ for animal waste (Kreitler 1975; Komor and Anderson 1993; and Kendall and Aravena 1999), but with some values ranging lower (Becker and others 2001 and Kendall, C. 1998).

In addition to animal waste, another potential source that the isotopic analysis can help determine is synthetic fertilizers. Unfortunately, the study did not evaluate the isotopic values for fertilizer, however. Given the lowest  $\delta^{15}\text{N-NH}_4$  value from the dairy lagoons was 2.0‰, it was decided to establish a value for  $\delta^{15}\text{N-NO}_3$  of 2.0‰ or less as the value in which isotopes could be used to identify synthetic fertilizer as the dominant source. The  $\delta^{15}\text{N-NO}_3$  value of 2.0‰ is generally supported from literature that suggests a range  $\delta^{15}\text{N-NO}_3$  values of -4.0 to +4.0‰ for synthetic fertilizers (Komor and Anderson 1993; Kendall, C.1998; and Kendall and Aravena 1999).

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For  $\delta^{15}\text{N-NO}_3$  values between 2.0‰ and 8.4‰, the source was identified as indeterminate, but with fertilizer and/or animal waste as potential sources of nitrate. This approach was taken because based on the data from this study, and the literature, it was not possible to determine the specific contribution from either fertilizer and/or animal waste for  $\delta^{15}\text{N-NO}_3$  values between 2.0‰ and 8.4‰.

**Commented [LE82]:** Do you really mean it could be all one or the other OR a combo? Because if you mean it is likely a combination, which is what I thought the text said, then "and/or" is confusing the issue. If you believe it is a combination, but that the proportions cannot be quantified, then it has to be "and."

The data for  $\delta^{18}\text{O-NO}_3$  was used to evaluate the degree to which an atmospheric signature was dominant in the sample. Values above  $\delta^{18}\text{O-NO}_3$  of 20.0‰ were considered "strong" contributors for the atmospherically derived nitrate component.  $\delta^{18}\text{O-NO}_3$  values below 20.0‰ could have an atmospheric contribution, but it was believed the contribution would be less.

Isotopic results were obtained from ammonium analyzed from the inlet to three sewer treatment plants in the Lower Yakima Valley. The plants were located in Zillah, Toppenish and Mabton and correspond to WWTP-01 through WWTP-03. The results for the analysis of  $\delta^{15}\text{N-NH}_4$  from the ammonium in the influent are presented below.

**Table B-2: Isotopic Analysis for WWTPs**

Location	$\delta^{15}\text{N-NH}_4$ (‰)	Assessment
WWTP-01: Zillah	3.72	Very Slightly Enriched
WWTP-02: Mabton	7.43	Some $\text{NH}_4$ Volatilization has occurred
WWTP-03: Toppenish	2.70	Very Slightly Enriched

$\delta^{15}\text{N-NH}_4$  values are expected to be lighter than the dairy wastes seen in the lagoon systems because of the limited opportunity for ammonia volatilization in the sewer system compared with the lagoon system. The value for the Toppenish system is comparable to the values seen in the dairy lagoons and may be a result of the more extensive system from this larger city. Values can be altered by other components such as materials added to the waste streams that cannot be controlled.

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## APPENDIX C – QUALITY ASSURANCE AND QUALITY CONTROL

This project was implemented in three phases. In Phase 1, a GIS screening application was developed and used to identify potential sample locations and sites in the Lower Yakima Valley for Phase 2 sampling and screening. Phase 1 also developed estimates of the relative nitrogen available for application to the land from different sources. Phase 2 and Phase 3 involved extensive sampling and analysis. A discussion of the quality assurance and quality control (QA/QC) followed in Phase 2 and Phase 3 is presented below.

Approximately 330 ~~residential~~ homes in the Yakima Valley were visited and tested for nitrates, ~~general~~ water quality parameters, and bacteria. As stated previously, the sampling teams used nitrate colorimetric test strips as a field screening tool to provide an indication of whether the water exceeded the EPA MCL for nitrate (10 mg/L). The Hach tests strips measure nitrate concentrations in increments of 0, 1, 2, 5, 10, 20, and 50 mg/L. If the test strip indicated the water ~~may might~~ exceed the MCL, samples were collected for analysis of nitrate by EPA's Manchester Environmental Laboratory. If there was any question whether the nitrate level was close to 10 mg/L, then the sampling team collected a sample for laboratory analysis. Table C-1 provides a summary of the field and laboratory measurements collected and analyzed and the corresponding project data quality goals.

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**Table C-1: Phase 2 - Field and Laboratory Measurements**

	Analytical Method	Reporting Limit	Accuracy Check	Precision Check
<b>Field Measurements</b>				
Temperature	170.1	0.5 C	+/- 0.5 C	+/- 1.5 C
Hydrogen Ion (pH)	150.1	0.1 unit	+/- 0.1 units	+/- 0.3 units
Dissolved Oxygen	360.1	0.3 mg/L	+/- 0.3 mg/L	+/- 0.5 mg/L
Specific Conductance	120.1	1 us/cm	+/- 7%	+/- 10%
Redox Potential	SM2580B	10 mv	+/- 0.5 mv	+/- 0.5 mv
Nitrate (Hach Test Strip)	Colorimetric	0.05 mg/L	+/- 0.5%	+/- 20%
<b>Laboratory Measurements</b>				
Nitrates	Method 300.0	0.06 mg/L	80-120%	+/- 20%
Chloride	Method 300.0	0.06 mg/L	80-120%	+/-20%
TKN	Method 351.2	0.5 mg/L	80 -120%	+/-20%

The field sampling team followed the QAPP specifications and adhered to the QA requirements set forth by the analytical protocols (EPA 2010a). A multi-parameter water quality instrument was used in the field for measuring dissolved oxygen, oxidation/reduction (redox) potential, pH, specific conductance, and temperature. All field instruments were calibrated prior to use. For quality control, duplicate sample readings, calibration checks, and matrix spike samples (if applicable) were performed. All field testing QC samples met the frequency of analysis.

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precision, and accuracy checks. Data generated are acceptable and can be used for screening purposes.

Based on the field screening test for nitrate, 102 water samples were shipped to EPA's Manchester Laboratory for analysis. Samples were confirmed for nitrates using EPA Method 300.0. Two percent of the total nitrate data points were qualified as follows: one sample (10086211) did not meet the holding time requirement and the result was qualified estimated; the nitrate concentration reported for this sample may be biased low. One sample (100866101) exceeded the highest level of the calibration curve and was qualified estimated. Data users are advised to consider the nitrate reported for this sample as biased low. All nitrates data, as reported and qualified, are acceptable for use for all purposes. All of the chloride and TKN analyses met the method required QC criteria. The data as reported are usable for all purposes.

Phase 2 was implemented following the specifications of the EPA approved "QA Project Plan for Yakima Nitrates Study, Phase 2 – Initial Nitrate/Coliform Screening of Domestic Wells, February 2010" (EPA 2010a). Deviations from the QAPP included changes in sample locations and modifications in the analytical method used, sampling method techniques, and additional number of samples collected. The rationale for these deviations was documented in the project team-approved Sample Alteration Form or Corrective Action Form.

In Phase 3, a new QAPP was developed and approved before Phase 3 sampling and analysis began (EPA 2010b). Based on Phase 2 screening results, samples and sub-samples were collected and shipped to the following laboratories for chemical analysis: EPA MEL, Cascade Analytical (Cascade), University of Nebraska Laboratory, USGS National Water Quality Laboratory (USGS-NWQL), USGS Laboratory in Reston (USGS-Reston) and EPA National Risk Management Laboratory RSKERC. Quality Assurance/Quality Control Summaries

#### 2.1 Manchester Environmental Laboratory, Port Orchard, Washington

A Stage 4 data validation was performed by the EPA Region 10 Quality Assurance (QA) team for all the data generated by MEL (**Appendix C Table 2**).

**Table C-2: Phase 3 - Summary of Chemical Analyses Conducted by MEL**

Matrix	# Samples	Parameter	Analytical Method (Prep)	Analytical Method (Analysis)
Water Wells, dairy lagoons, and WWTP influent	49	TKN	AOAC 933.13	
	49	NH3	SM 10-107 (04-1-A version)	
	49	Nitrates-Nitrites	EPA Method 351.2	
	49	Total Metals	EPA Method 200.2	EPA Method 200.7
	49	Mercury	EPA Method 245.1	
	49	Alkalinity	Method 2320B	
	31	Coliforms	9221F, 9221E, 9222B	

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**Table C-2: Phase 3 - Summary of Chemical Analyses Conducted by MEL**

Matrix	# Samples	Parameter	Analytical Method (Prep)	Analytical Method (Analysis)
	13	Microbial Source Tracking	DNA PCR Techniques	
	49	Bromide, Chloride, Fluoride, and Sulfate	EPA Method 300.0	
	49	Total Phosphorous	EPA Method 365.1	
	30	Pesticides (only water wells)	Method 551.1	SW846 – Method 8270D-SIM
	30	Herbicides (only water wells)	SW846-Method 8151A	SW846-Method 8270D-SIM
Soils and Manure	16	Mercury	EPA 7471	
	16	Alkalinity	Method 2320B	
	16	Total Metals	EPA Method 200.2	EPA Method 200.7
	16	Pesticides	Method 551.1	SW846 – Method 8270D-SIM
	16	Herbicides	SW846-Method 8151A	SW846-Method 8270D-SIM

All of the chemical and microbial analyses conducted at MEL met the project data quality goals and criteria for accuracy, precision, comparability, completion, representativeness, and sensitivity, and are useable for all purposes with the following exceptions:

Nitrates and Nitrogen Compounds

Nitrogen compounds included ammonia, TKN, and nitrates-nitrites. Samples 10154251, 10154252, 10154253, 10154254, 10154255, 10154256, 10154257, 10154258, 10154259, 10164260, 10164261, 10164262, 10164263, 10164264 and 10164265 did not meet the required preservation when they were received at the laboratory. Nitrates/nitrites and ammonia results for these samples were qualified estimated with a possible low bias. Nineteen percent of the total data points (147) were qualified.

Mercury and Alkalinity

Approximately 37 percent of the total mercury data points were qualified estimated based on out of control sample spike and blank spike recoveries. Alkalinity results met all the QC criteria. The mercury and alkalinity data, as reported and qualified, are acceptable for use for all purposes.

#### Pesticides and Herbicides

The project data quality goals for precision and accuracy for numerous target analytes were not met for dairy lagoons and WWTPs. As stated above, all of the pesticides and herbicide results for the dairy lagoons and WWTPs could not be quantified and are considered unusable because of (1) the complexity of the sample matrices, (2) holding times that were exceeded, (3) recurring QC failures, and (4) the limitations of modified Method 8270D for detecting pesticides and herbicides at the project reporting levels. However, the pesticides for water and soil, as qualified, are usable for all purposes.

#### Anions

Anions included chloride, fluoride, bromide, and sulfates. As a result of matrix interferences, the dairy lagoon and WWTP biosolids samples collected were analyzed at 50x dilutions for bromide, fluoride, and sulfate. The reporting limits for these bromide, fluoride, and sulfate were elevated and did not meet the project goals. As qualified and reported, the analytical results for water and soil are acceptable for use for all purposes.

### 3.2 Cascade Analytical Laboratory, Wenatchee, Washington

#### Nitrate and Other Forms of Nitrogen

Cascade Analytical Laboratory is a is certified by the State of Washington to conducted drinking water analysis including analysis for nitrate. It is located in Union Gap and Wenatchee Washington, and analyzed nitrate for this study. Due to the short holding times for certain nitrates analytical methods, Cascade Analytical Laboratory was sub-contracted by Region 10 to analyze the water well, soil, and manure samples for nitrate and nitrogen compounds for Phase 3. A total of 30 water wells, 11 soil, and five manure samples were submitted.

The analytical method used for the determination of nitrates in water samples was Method 300.0. The methods used for the analysis of total nitrogen/solid, ammonia solid, and nitrate-nitrite in solid and manure samples were the Association of Analytical Communities (AOAC) Method 993.13, Standard Method 4500-NH3, and Method 4500-NO3. A Stage 4 data validation was performed by EPA Region 10 QA team for all data generated by Cascade Analytical Laboratory.

All of the QC samples and sample analysis met the technical acceptance criteria set forth by the methods. The data, as reported, are acceptable for use for all purposes.

Thirty split water samples were collected, shipped to Cascade Analytical Laboratory and MEL, and analyzed for nitrates using the EPA Method 300.0 (Cascade) and EPA Method 351.2 (MEL). Both sets of data met all the method-specified QC criteria and are acceptable for use for all purposes. The nitrates concentrations reported by both laboratories are comparable within 10 percent. The following is a list of water samples that were collected, split, and sent to these two labs:

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10154201	10154202	10154203	10154204	10154205
10154206	10154207	10154208	10154211	10154212
10154213	10154214	10154215	10154216	10154217
10154218	10154219	10154220	10154221	10154223
10154224	10154225	10154226	10154227	10154228
10154229	10164209	10164210	10164222	10164230

#### **4.3 EPA National Risk Management Research Laboratory, Robert S. Kerr Environmental Research Center, Ada, OKRSKERC**

##### Hormone and Perchlorate Analyses

Fifteen dairy lagoon, three WWTP, and 30 water samples were analyzed for estrogens (17- $\alpha$ -estradiol; 17- $\beta$ -estradiol; 17- $\alpha$ -ethynyl estradiol; estriol; and estrone) by EPA's Robert S. Kerr Environmental Research Center following the in-house standard operating procedure (SOP) "*Quantitation of Estrogens in Groundwater and Animal Waste Dairy lagoon Water Using Solid Phase Extraction, Pentafluorobenzyl and Trimethylsilyl Derivatization and Gas Chromatography Negative Ion Chemical Ionization/Mass Spectrometry/Mass Spectrometry, RSKSOP-253, Revision 2, October 2010.*"

The same 30 water samples were also analyzed for perchlorate following the modified *USEPA SW846 Method 6850, "Perchlorate in Soils, Water and Wastes Using High performance Liquid Chromatography/Electrospray/Ionization (ESI) Mass Spectroscopy (MS) or Tandem Mass Spectroscopy (MS/MS)*. All sample analyses were evaluated following the EPA's Stage 2B Manual Data Validation Process. The summaries of sample and QC analyses were evaluated and laboratory qualifiers were mapped to Region 10 EPA validation qualifiers following the technical acceptance criteria and method quality control specifications. All of the technical acceptance criteria for QC were met by both analyses. Target compounds detected above the Method Detection Limit (MDL) but below reporting limits were qualified estimated, "J." Data detected below the MDL were qualified non-detects, "U," and reported at the MDL level. The data, as qualified, are usable for all purposes.

#### **5.4 USGS National Water Quality Laboratory, Denver, Colorado**

##### Trace Organics

Fifteen dairy lagoons, three WWTP plant influent, and 30 water samples were analyzed for trace organic chemicals following the SOP for the "*Analysis of Waste Water Samples by Gas Chromatography/Mass Spectroscopy*" – USGS SOPs 1433 and 4433. All sample analyses were evaluated following EPA's Stage 2B Manual Data Validation Process (S2VM). The summaries of sample and QC analyses were evaluated and laboratory qualifiers were mapped to Region 10 EPA validation qualifiers following the technical acceptance criteria and method quality control

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specifications. Samples were analyzed following the technical specifications of the analytical method.

The data, as qualified, are usable for all purposes except for approximately 6 percent of the total data points, which were qualified unusable based on extremely low surrogate recoveries. Approximately 32 percent of the total data points were qualified estimated as a result of chromatographic interference and QC results that did not meet the specified criteria.

Trace levels of 4-tert-octylphenol, diethyl phthalate, menthol, p-cresol, tri(2-butoxyethyl)phosphate, tri(2-chloroethyl) phosphate, tri (dichloroisopropyl) phosphate, tributyl phosphate, and triphenyl phosphate were detected in the field blank (WW29). Only the diethyl phthalate in associated sample WW06 detected at a concentration less than 5x the value in the blank was qualified as non-detect, "U," based on blank contamination.

#### University of Nebraska – Water Science Laboratory, Lincoln, Nebraska (UNL)

The University of Nebraska analyzed several different types of compounds. **Table C-3** provides a summary of the compounds evaluated, number of samples, matrix, and analytical method.

**C-3: Phase 3 - Summary of Analyses Conducted by UNL**

Matrix	Compounds	No. of Samples	Analytical Method (Prep)	Analytical Method (Analysis)
Water Wells, dairy lagoons, and WWTP influent	Hormones	47	On-line SPE with C18 clean-up	SOP#LCMS_APPI_Steroids_Water-001
	Waste water contaminants	47	Off-line SPE-Modified Method 3535	LC/MS SOP-LCQ-Wastewater-001
	Pharmaceuticals	47	On-line SPE extraction with citrate buffer	SOP#LC/MS_Vet_P harm_water-002
	Isotopic Nitrogen	47	Analyte Prep 15-002	N15 Analysis Dual Inlet IRMS
	Isotopic Oxygen	47	SOP#Analyte-O18 in Nitrate/AgNO3	SOP# Inst-Isoprime EA-18O-001
	Ammonia	47	Analyte-DISTN15-004	Titrimetric
	Nitrate	47	Analyte Prep 15-002	Titrimetric
Soil and Manure	Hormones	16	Microwave-Assisted solvent extraction (MASE) and SPE	SOP# Analyte-Steroids_Solids-001
	Waste water contaminants	16	Microwave-Assisted solvent extraction (MASE) and SPE	SOP#-Analyte-LCQ-Wastesolid-001
	Pharmaceuticals	16	On-line SPE extraction with citrate buffer	SOP#-Analyte-VetPharmSED-001

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A Stage 2A data validation review was conducted by the EPA QA team on all the data. The validation included the limited evaluation of calibration, QA, and sample analytical summary results. All samples were analyzed following the technical specifications of UNL's in-house SOPs.

#### General QA Observations for UNL Analyses

UNL data sets may not meet the third-party reproducibility criterion set forth by EPA's Information Quality Guidelines (EPA /260R-02-008 October 2002) for the following reasons: (1) there is no established or standard analytical method for the analysis of the target compounds, and the analytical methods used are for research purposes only, (2) the recurrence of out-of-control QC results; (3) variability in duplicate runs; and (4) compound identification and calculations were not verified at the time of review because the instruments' raw data output was not available.

Twenty-nine water, 15 dairy lagoons, three WWTP, and 16 soil or manure samples were collected and analyzed for wastewater pharmaceuticals, veterinary pharmaceuticals, hormones and steroids, and isotopic nitrogen and isotopic oxygen. The following is a summary of the data validations for UNL:

Wastewater Pharmaceuticals: Approximately 10 percent of the wastewater pharmaceutical data points were qualified unusable because of extremely low spike and surrogate recoveries (less than 10 percent). The rest of the data as qualified are usable for all purposes.

Veterinary Pharmaceuticals: No significant problems were encountered with the analysis of soil/solid samples for veterinary pharmaceuticals. Most of the liquid samples (dairy lagoons, well water, and WWTP) underwent multiple analyses because of concentrations of some of the target compounds in the field blank and also because of matrix interferences. Approximately 9 percent of the total data points were qualified unusable and an additional 18 percent were qualified estimated concentrations with a high bias because of out of control internal standards or calibration. Five lincomycin and three monensin results in the water samples were detected above the reporting limits but were flagged non-detects based on contamination in the associated field blank, WW29. The concentrations reported were calculated using internal standard techniques. Most of the internal standards did not meet minimum area requirements when compared with the daily calibration standards. Therefore, the associated results may be biased high.

Steroids/Hormones: Because of the calibration results, the detected results or reporting limits for androstenedione, androsterone, progesterone, estrone, a-zearalanol, a-zearalenol, b-zearalanol for samples associated with the calibration run on January 18, 2011, were qualified estimated, "J/UJ." Approximately 15 percent of the total data points were flagged estimated because of calibrations. In addition, some target compounds were qualified non-detects based on contamination in the associated blank.

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Isotopic Nitrogen/Isotopic Oxygen Analyses/Ammonium and Nitrate Nitrogen Analyses: Isotopic nitrogen and oxygen were determined using the amounts of ammonium and nitrate-nitrogen in water. No problems were encountered with the isotopic nitrogen, isotopic oxygen, and intermediate ammonia and nitrates nitrogen results. For QC, laboratory reagent blanks, duplicates, and laboratory-fortified blanks were analyzed at the required frequency. All of the results were comparable to each other. Data were not qualified and usable for all purposes.

#### 7.6 USGS Laboratory, Reston Virginia

##### Recharge Age Dating

The USGS Laboratory located in Reston analyzed the recharge age of the water well samples following the SF<sub>6</sub> procedure.

Limitations of the Method: The recharge dating procedure is a statistical calculation derived from the SF<sub>6</sub> gas evolved in the sample and other existing data. It is applicable to young groundwater systems aged 1970 to present. This procedure is not applicable to areas with high anthropogenic and natural SF<sub>6</sub> background values such as indicated by samples WW-01, WW-11, WW-12, WW-13, WW-23, and WW-28. As a result, age could not be measured in those samples because of the high values of SF<sub>6</sub> as dissolved gases from the sample. These samples may indicate areas where localized anthropogenic sources of SF<sub>6</sub> exist. Alternatively, volcanic rocks can contain more SF<sub>6</sub> than the average atmospheric concentrations of SF<sub>6</sub> and the volcanic terrain and mineralogy of the sediments in the local aquifer may be the source of the SF<sub>6</sub>.

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The USGS laboratory flagged these six water wells samples with a "C" qualifier, meaning contaminated. For clarity, the validator changed the "C" qualifier with "NM," not measured. In addition, there were also some samples with recharge calculated dates before 1970. The dating technique used provides only a range, and data users should be warned that the reported recharge ages are estimates.

## APPENDIX D: INFORMATION ON THE R&M Haak Dairy

The R&M Haak Dairy (Dairy) is located in an agricultural area north of the Yakima River, about four miles north of the city of Sunnyside. It is in the Benton groundwater basin, which includes the communities of Sunnyside, Grandview, Satus, Kiona, Prosser, Mabton and Richland (Jones and others 2006). This Dairy was selected as one of the sampling locations because it is relatively high on the landscape with very few other sources of nitrate above the dairy. The Dairy A ditch runs from north to south through the Dairy. Cow pens, a milking parlor, and three waste lagoons lie west of the ditch. There are several large structures where cows are kept. East of the ditch, a center-pivot irrigation system is installed on a large sprayfield which is used by the Dairy. The Dairy operator stated that corn and triticale were alternately grown on the sprayfield. See Appendix D for a more detailed description of the Haak Dairy and its operations.

Within the Haak dairy property boundary, five soil units have been mapped by the U.S. National Resource Conservation Service (NRCS, 2011). All five soil units have a silt loam texture with a "well-drained" classification. Three of the soil units (Scooteney, Sinloc, and Warden) represent 82% of the surface area. They have a saturated hydraulic in the range of 1.1 to 4.0 feet per day, which is characterized as "moderately high to high" in their capacity to transmit water.<sup>3</sup> Likewise the other two soil units (Burke and Scoon) have a moderately high to high capacity to transmit water to a depth of 2 to 3 feet bgs; however, below this depth a cemented layer is present with a saturated hydraulic conductivity in the "very low to moderately low" range of 0.00 to 0.12 feet per day. The Burke and Scoon map units are located in the northwest portion of the Haak dairy property and represent 18% of the surface area.

The NRCS information shows the surface of the Haak sprayfield, which is irrigated with nitrogenous liquid Dairy wastewater from the lagoons, consists almost entirely of highly permeable soils. Water from irrigation or precipitation can carry nitrogen through the surface soils at a rate of 1.1 to 4.0 feet per day. Wastewater applied onto these highly permeable soils increases the risk that nitrogen will be transported downward to the drinking water aquifer before it can be taken up by plants, even if the plants have the capacity to take up more. Elevated levels of nitrogen in soils can lead to groundwater contamination, especially if these conditions exist on soils of relatively high permeability. The capacity of most of the surface soils within the Haak property boundary (about 82% of the surface area) to transmit water can be characterized as "moderately high to high".

### Waste management at the Haak Dairy

Dairy inspection reports obtained from the Washington Department of Agriculture indicate the Haak Dairy uses a lagoon system with a capacity of 9.4 million gallons. The surface area of the first lagoon is approximately 70,000 square feet and is roughly triangular in shape. The second lagoon is roughly rectangular and is about 37,000 square feet, and the third is roughly triangular

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<sup>3</sup> Saturated hydraulic conductivity amounts have been converted from inches per hour to feet per day.

and is about 162,000 square feet. The lagoon system was 80 percent full at the time of the July 2008 inspection and 75 percent full at the time of the March 2010 inspection.

In addition to the lagoons, the Haak Dairy employs a solids separator and composting to treat waste. Animal corpses are composted. Waste is applied to crop fields by spreader ("honey wagon"), a sprinkler irrigation system, and a dry spreader. The inspection reports indicate the waste from the dairy was applied to six fields at the time of the inspections.

#### Potential sources of Nitrogen sampled

EPA sampled several likely sources of nitrogen on the Haak Dairy. Composited samples were taken from a large manure pile, and from the dairy sprayfield. EPA also sampled liquid effluent from the lagoon system.

A manure pile can be a source of nitrogen to the groundwater if it becomes wet. The manure pile sampled at the Haak Dairy was on bare ground just south of one of the structures where the cows were kept and not covered. The manure had high moisture content and appeared to have been recently pushed off of a concrete pad onto the ground. A composite sample was taken to provide a representative sample of the pile.

Using the seepage rate range of 0.2 to 2.4 mm/day observed by Ham and DeSutter, a lagoon system with a similar surface area to the one at Haak would be expected to have a leakage rate in the range of 482,000 gallons (roughly the volume of 0.7 Olympic-size swimming pools) to 5,783,000 gallons (roughly 8.8 pools) of liquid waste per year into the underlying soil.

EPA took a composite soil sample of a large crop field irrigated with animal waste from the lagoon system. The sample results shows a total nitrogen level of 2,760 part per million.

#### Other possible sources of nitrogen from the Haak Dairy

Synthetic inorganic fertilizer is a potential source of nitrogen from the Haak Dairy. Dairy inspection reports indicate that the dairy has used commercial fertilizer in the recent years. Cow manure contains several plant nutrients including nitrogen and phosphorus. Phosphorus is often the limiting nutrient in how much manure can be applied because the amount of manure should be tailored to the nutrient needs of the type of crop being grown, and manure has a larger proportion of phosphorus relative to nitrogen for most crop needs. Applying too much phosphorus can cause surface water pollution. Growers sometimes apply manure until the phosphorus need is met and make up for the nitrogen shortfall by supplementing with inorganic nitrogen fertilizer. The report does not indicate whether the fertilizer used by the Haak Dairy contained nitrogen.

Because the area around the Haak Dairy is not served by a municipal sanitary sewer system, the dairy presumably provides an on-site septic system for its employees. The detection of human fecal bacteria in the dairy lagoons suggests that waste from the facility's septic system has

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somehow made its way into the lagoons. Liquid leaking from the lagoons into the subsurface  
soils could contain nitrogen from both animal and human sources.

#### **APPENDIX D REFERENCES**

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**APPENDIX E: INFORMATION ON DAIRY CLUSTER (In the process of  
being developed)**

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# Source and transport controls on the movement of nitrate to public supply wells in selected principal aquifers of the United States

P. B. McMahon,<sup>1</sup> J. K. Böhlke,<sup>2</sup> L. J. Kauffman,<sup>3</sup> K. L. Kipp,<sup>1</sup> M. K. Landon,<sup>4</sup> C. A. Crandall,<sup>5</sup> K. R. Burow,<sup>6</sup> and C. J. Brown<sup>7</sup>

Received 12 June 2007; revised 29 October 2007; accepted 7 December 2007; published 4 April 2008.

[1] In 2003–2005, systematic studies in four contrasting hydrogeologic settings were undertaken to improve understanding of source and transport controls on nitrate movement to public supply wells (PSW) in principal aquifers of the United States. Chemical, isotopic, and age tracer data show that agricultural fertilizers and urban septic leachate were the primary sources of large nitrate concentrations in PSW capture zones at Modesto, California (Central Valley aquifer system) and York, Nebraska (High Plains aquifer). Urban septic leachate and fertilizer (possibly nonfarm) were the primary sources of large nitrate concentrations in PSW capture zones at Woodbury, Connecticut (glacial aquifer system), and Tampa, Florida (Floridan aquifer system), respectively. Nitrate fluxes to the water table were larger in agricultural settings than urban settings, indicating that it would be beneficial to reduce PSW capture zone areas in agricultural regions. Mixing calculations indicate that about 50 to 85% of the nitrate in water from the PSW could be from those modern anthropogenic sources, with the remainder coming from sources in old (>50 years) recharge or sources in young recharge in undisturbed settings such as forests. Excess  $N_2$  concentrations and age tracers showed that denitrification at Modesto occurred gradually (first-order rate constant of 0.02/a) in a thick reaction zone following a ~30-year lag time after recharge. Denitrification generally was not an important nitrate sink at Woodbury. At York and Tampa, denitrification occurred rapidly (0.5 to 6/a) in thin reaction zones in fine-grained sediments that separated the anoxic PSW producing zones from overlying oxic, high-nitrate ground water. Particle tracking showed that a major pathway by which anthropogenic nitrate reached the York and Tampa PSW was by movement through long well screens crossing multiple hydrogeologic units (York) and by movement through karst features (Tampa), processes which reduced ground water residence times in the denitrifying zones. These results illustrate how PSW vulnerability to nitrate contamination depends on complex variations and interactions between contaminant sources, reaction rates, transit times, mixing, and perturbation of ground water flow in contrasting hydrogeologic settings.

**Citation:** McMahon, P. B., J. K. Böhlke, L. J. Kauffman, K. L. Kipp, M. K. Landon, C. A. Crandall, K. R. Burow, and C. J. Brown (2008), Source and transport controls on the movement of nitrate to public supply wells in selected principal aquifers of the United States, *Water Resour. Res.*, 44, W04401, doi:10.1029/2007WR006252.

## 1. Introduction

[2] Nitrate ( $NO_3^-$ ) is one of the most common ground water contaminants in the world and its presence in the environment at elevated concentrations poses well-known human health and ecological risks [Fan and Steinberg, 1996; Galloway *et al.*, 2003]. In 2000, about 37% of the public water supply in the United States came from ground

water [Hutson *et al.*, 2004]. A nationwide survey of  $NO_3^-$ , pesticides, and volatile organic compounds in untreated water from domestic and public supply wells (PSW) in the United States showed that  $NO_3^-$  was the contaminant that most frequently exceeded a federal drinking water standard (10 mg/L as N for  $NO_3^-$ ) [Squillace *et al.*, 2002]. Similarly, the U.S. Environmental Protection Agency reported that among organic and inorganic contaminants in all types of public water systems,  $NO_3^-$  was the one that most frequently exceeded a federal drinking water standard [U.S. Environmental Protection Agency, 2005]. In California, about 10% of the 38,000 water samples collected from PSW exceeded the  $NO_3^-$  drinking water standard in a 1975 to 1987 survey [Franco *et al.*, 1994]. Despite the importance of ground water to the nation's water supply and the relative impact of  $NO_3^-$  on the quality of that resource, detailed and systematic assessments of the processes re-

<sup>1</sup>U.S. Geological Survey, Denver, Colorado, USA.

<sup>2</sup>U.S. Geological Survey, Reston, Virginia, USA.

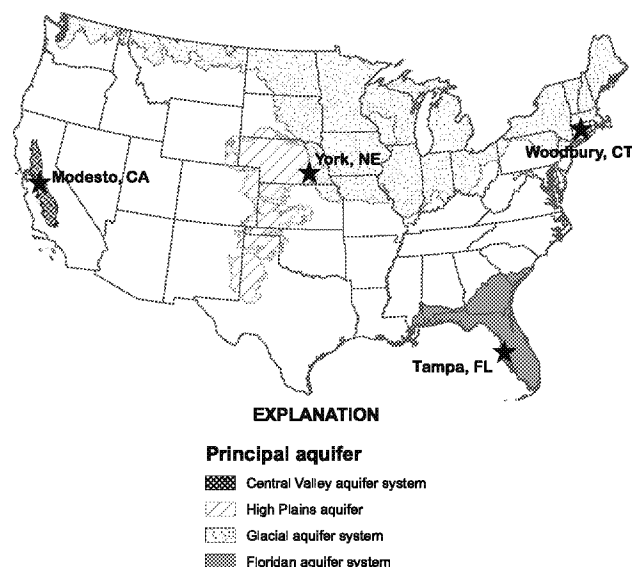
<sup>3</sup>U.S. Geological Survey, West Trenton, New Jersey, USA.

<sup>4</sup>U.S. Geological Survey, San Diego, California, USA.

<sup>5</sup>U.S. Geological Survey, Tallahassee, Florida, USA.

<sup>6</sup>U.S. Geological Survey, Sacramento, California, USA.

<sup>7</sup>U.S. Geological Survey, Hartford, Connecticut, USA.



**Figure 1.** Location of the principal aquifers and communities where public supply wells were studied.

sponsible for  $\text{NO}_3^-$  contamination in PSW generally are lacking [Kauffman *et al.*, 2001].

[3] Understanding the origin, fate, and transport of  $\text{NO}_3^-$  and other contaminants to PSW is challenging for a number of reasons. Large-capacity pumping wells could have large and complex areas contributing recharge to the well screens [Barlow, 1997; Kauffman *et al.*, 2001], in which the land use (contaminant source) varies temporally and spatially. Under nonpumping conditions, water movement across long-screened intervals common in PSW could vertically redistribute contaminants in the flow system [Reilly *et al.*, 1989; Shapiro, 2002; Konikow and Hornberger, 2006]. Under pumping conditions, composite samples from long-screened intervals could consist of waters of varying ages and chemical compositions [Reilly and LeBlanc, 1998; Weissmann *et al.*, 2002], thus complicating interpretations of contaminant source and fate. Studies of contaminant movement to PSW that address these and other related issues could lead to better understanding of how to locate, construct, and operate PSW to minimize contamination. In 2001, the U.S. Geological Survey began a systematic assessment of processes affecting the transport of anthropogenic and natural contaminants, including  $\text{NO}_3^-$ , to PSW in the United States [Eberts *et al.*, 2005]. The purpose of this report is to examine source and transport controls on the movement of  $\text{NO}_3^-$  to PSW in selected principal aquifers of the United States.

[4] A single PSW in each of four communities across the United States was selected for detailed study. The communities and the principal aquifers in which the wells are screened are Modesto, California (Central Valley aquifer system); York, Nebraska (High Plains aquifer); Woodbury, Connecticut (Glacial aquifer system); and Tampa, Florida (Floridan aquifer system) (Figure 1). These four communities and principal aquifers were selected to represent a range of land uses, population sizes, and hydrogeologic conditions. In each area, ground water was a major source, if not the sole source, of water supply. Similar monitoring well network designs, sampling methodologies, broad suites of

chemical and isotopic parameters, and analytical and modeling methods were used at all sites to enhance comparability of results between sites. Thus, this study represents one of the first to systematically evaluate source and transport controls on  $\text{NO}_3^-$  movement to PSW in diverse hydrogeologic settings.

## 2. Study Area Descriptions

### 2.1. Modesto, California

[5] Modesto, with a population of 188,856 (U.S. Census Bureau, 2000, U.S. Census 2000 American Fact Finder, accessed 13 April 2006, available at [http://factfinder.census.gov/home/saff/main.html?\\_lang=en](http://factfinder.census.gov/home/saff/main.html?_lang=en)), is located in the Central Valley of California. The climate is semiarid and the area receives an average of 315 mm of precipitation each year. The Central Valley is a northwest trending structural basin bounded on the west by the Coast Range and on the east by the Sierra Nevada. Sediments in the study area largely consist of a series of overlapping terrace and alluvial fan deposits derived from the Sierra Nevada. Unconsolidated clay, sand, and gravels of the Pliocene-Pleistocene aged Laguna, Turlock Lake, and Riverbank Formations constitute the Central Valley aquifer in the study area (Figure 2). The aquifer in the study area is at least 100 m thick, and the depth to water was about 10 m. Characteristics of the PSW selected for study and land use in the area contributing recharge to the well are listed in Table 1. A map showing the area contributing recharge to the Modesto PSW is presented in Figure S1<sup>1</sup>. Recharge rates in the agricultural and urban areas were about 400–700 and 270 mm/a, respectively (Table 2). The primary sources of recharge are precipitation and irrigation return flow.

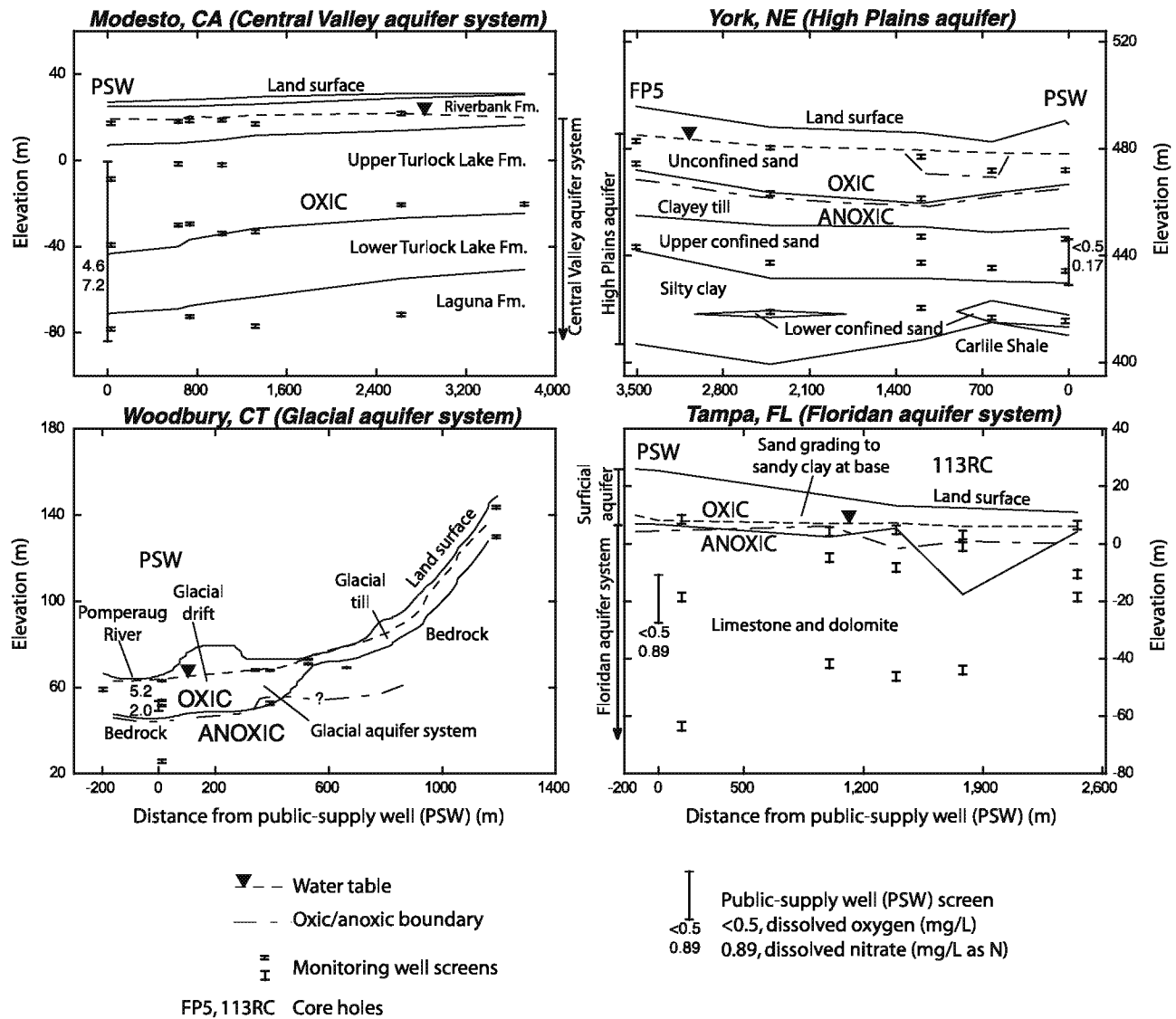
### 2.2. York, Nebraska

[6] York, with a population of 8081 (U.S. Census Bureau, 2000), is located within the High Plains in eastern Nebraska. The area has a humid, continental climate and receives an average of 711 mm of precipitation each year. The High Plains in eastern Nebraska is underlain by heterogeneous deposits of gravel, sand, silt, and clay of Quaternary age that form a layered sequence of coarse- and fine-grained units. A laterally continuous clayey till deposit 8 to 17 m in thickness separates upper unconfined coarse sands and lower confined fine sands of glaciofluvial origin (Figure 2). These units represent the High Plains aquifer in the study area. These unconsolidated sediments are underlain by the Carlisle Shale, which is marine shale of Cretaceous age. The High Plains aquifer in the study area is about 80 m thick, and the depth to water was about 10 m. Characteristics of the PSW selected for study and land use in the area contributing recharge to the well are listed in Table 1. The area contributing recharge is shown in Figure S1. Recharge rates in the agricultural and urban areas were about 210 and 70 mm/a, respectively (Table 2). The primary sources of recharge are precipitation and irrigation return flow.

### 2.3. Woodbury, Connecticut

[7] Woodbury, with a population of 9198 (U.S. Census Bureau, 2000), is located within the glaciated New England

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2007WR006252.



**Figure 2.** Hydrogeologic sections in the study areas. Oxidic waters are operationally defined by dissolved oxygen  $\geq 0.5$  mg/L. See Figure S1 for map location of the sections.

Uplands in west central Connecticut. The area has a humid continental climate and receives an average of 1170 mm of precipitation each year. Near-surface sedimentary deposits in the Pomperaug River valley consist of stratified glacial drift in the river valley and glacial till deposits on the valley sides (Figure 2). These units are part of the North-eastern Glacial aquifer system. The valley is underlain by Mesozoic sedimentary and Paleozoic crystalline rocks. The valley sides and ridges are underlain by Paleozoic rocks that include granite, quartzite, schist, and gneiss. The Glacial aquifer in the study area is about 23 m thick, and the depth to water was about 4 m. Characteristics of the PSW selected for study and land use in the area contributing recharge to the well are listed in Table 1. The area contributing recharge is shown in Figure S1. The recharge rate in the study area was about 650 mm/a (Table 2). The primary sources of recharge are precipitation and infiltration of streamflow.

## 2.4. Tampa, Florida

[8] Tampa, with a population of 303,447 (U.S. Census Bureau, 2000), is located on Florida's Gulf Coast. The area has a subtropical climate and receives an average of 1,140 mm of precipitation each year. The study area is underlain by a thick sequence of limestone and dolomite of Eocene through Miocene age that is characterized by well-developed karst features such as springs, conduits, and sinkholes (Figure 2). These rocks constitute the Floridan aquifer in the study area. They are overlain by a discontinuous layer of late Miocene sediments composed of dense, plastic, green-gray clay, interbedded with varying amounts of chert, sand, clay, marl, shell, and phosphate. In places, this clay layer was breached by sinkholes associated with the underlying carbonate rocks. Overlying those sediments are unconsolidated sands, clays, and marls of Pliocene to Holocene age that constitute the Surficial aquifer. Depth to water in this aquifer generally was within 3 m of land surface. The Floridan aquifer in the study area is at least



**Table 1.** Characteristics of the Public Supply Wells Selected for Study

Location	County	Year of Construction	Average Pumping Rate (m <sup>3</sup> /d)	Screened Interval (m below land surface)	Area Contributing Recharge (km <sup>2</sup> ) <sup>b</sup>	Generalized Land Use in Area Contributing Recharge (%) <sup>a</sup>				
						Agriculture	Urban	Forest	Wetland	Rangeland
Modesto, California	Stanislaus	1961	3750	28 to 111	4.2	30	67	<1	<1	2.3
York, Nebraska	York	1977	1390	43 to 61	6.3	39	45	11	<1	3.7
Woodbury, Connecticut	Litchfield	1967	390	14 to 18	0.5	4.8	75	13	6.9	<1
Tampa, Florida	Hillsborough	1958	2690	36 to 53	2.3	4.2	84	1	5.0	5.8

<sup>a</sup>On the basis of 1992 National Land Cover Data (NLCD) [Vogelmann *et al.*, 2001].

<sup>b</sup>Areas contributing recharge were derived from particle tracking models.

100 m thick. Characteristics of the PSW selected for study and land use in the area contributing recharge to the well are listed in Table 1. The area contributing recharge is shown in Figure S1. The recharge rate in the urban area was about 380 mm/a (Table 2). The primary sources of recharge are precipitation and infiltration of stream flow.

### 3. Methods

#### 3.1. Particle Pathline Analysis

[9] Ground water pathlines and advective travel times were calculated using MODPATH [Pollock, 1994] to delineate the areas contributing recharge to the PSW, identify approximate flow paths to the PSW, and estimate age distributions of water captured by the PSW. Existing steady state, three-dimensional ground water flow models done using MODFLOW-2000 [Harbaugh *et al.*, 2000; Starn and Brown, 2007; Clark *et al.*, 2007; Birow *et al.*, 2008; U.S. Geological Survey, unpublished data, 2007] provided input into MODPATH. The forward tracking option of MODPATH was used for pathline analysis. From 2,344 to 47,008 particles were started at all inflow locations in the models and tracked to their discharge point. The particles were uniformly distributed on the face of the model cells associated with inflow to the model. Particles captured by the PSW were used to delineate areas contributing recharge and flow paths to those wells. The flow associated with each particle was computed by dividing the inflow at the source face by the number of particles started at that face. Effective

porosity is required for pathline analysis and assigned values ranged from 0.2 to 0.45. Maps of the three-dimensional distribution of redox zones in the flow systems were superimposed on the pathlines to determine travel times in each zone.

#### 3.2. Sampling

[10] Water table monitoring wells were installed under the major land uses in or near the contributing areas to characterize the chemical composition of recent recharge in each land use (Figure S1). Nested monitoring wells were installed along the MODPATH identified flow paths to the PSW to study the history, transport, and fate of NO<sub>3</sub><sup>-</sup>. At the PSW, composite water samples were collected at the well-head and depth-dependent samples were collected from discrete depth intervals in the well screen during pumping. Water samples were analyzed for over 100 chemical and isotopic parameters but only dissolved ammonium (NH<sub>4</sub><sup>+</sup>), nitrite (NO<sub>2</sub><sup>-</sup>), and NO<sub>3</sub><sup>-</sup>; dissolved gases (Ne, Ar, O<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>); multiple isotopes (<sup>3</sup>H,  $\delta^{15}\text{N}[\text{NO}_3^-]$ ,  $\delta^{18}\text{O}[\text{NO}_3^-]$ ,  $\delta^{15}\text{N}[\text{N}_2]$ ); and age tracers (CFC-11, CFC-12, CFC-113, SF<sub>6</sub>, <sup>3</sup>H/<sup>3</sup>He) are reported here. Further details on the collection and analysis of water samples are provided in Text S1.

#### 3.3. Modeling Oxygen Reduction and Denitrification Rates

[11] Different approaches were used to estimate rates of O<sub>2</sub> reduction and denitrification (microbial reduction of NO<sub>3</sub><sup>-</sup> to N<sub>2</sub>), depending on the spatial and temporal scales

**Table 2.** Median NO<sub>3</sub><sup>-</sup> Concentration in Recharge and Median NO<sub>3</sub><sup>-</sup> Flux to the Water Table in Areas Contributing Recharge to the Public Supply Wells<sup>a</sup>

Location	Recharge Rate (mm/a)		NO <sub>3</sub> <sup>-</sup> Concentration in Recharge (mg/L as N)		NO <sub>3</sub> <sup>-</sup> Flux to Water Table (kg N/ha-year)		Probable Source of Large NO <sub>3</sub> <sup>-</sup> Concentrations in Ground Water	
	Agriculture	Urban	Agriculture	Urban	Agriculture	Urban	Agriculture	Urban
Modesto, California	400 to 700	270	12.5 (3.98 to 20.6)	3.48 (2.06 to 11.1)	69 (16 to 144)	9.4 (5.6 to 30)	Fertilizer	Septic Leachate, Fertilizer
York, Nebraska	210	70	15.7 (5.21 to 77.9)	22.5 (15.9 to 28.8)	33 (11 to 169)	16 (11 to 20)	Fertilizer, Manure spreading	Septic Leachate <sup>b</sup>
Woodbury, Connecticut	Not applicable	650	Not applicable	1.14 (<0.06 to 2.23)	Not applicable	7.4 (<0.4 to 14)	Not applicable	Septic Leachate, Fertilizer
Tampa, Florida	Not applicable	380	Not applicable	2.45 (0.21 to 4.56)	Not applicable	9.3 (0.8 to 17)	Not applicable	Fertilizer

<sup>a</sup>Ranges of values are reported in parentheses. Probable sources of large NO<sub>3</sub><sup>-</sup> concentrations in ground water were selected on the basis of <sup>15</sup>N values, chemical data, and land use information.

<sup>b</sup>Septic leachate could include leaking sewer lines.

at which reactions occurred and on the density of sampling points in the reaction zones. *Groffman et al.* [2006] discuss in more detail scale issues associated with estimating in situ denitrification rates. In thick reaction zones where  $O_2$  reduction and denitrification appeared to occur gradually,  $O_2$  reduction and denitrification rates were estimated by comparing gradients in reaction progress and ground water age using the following models:

$$-R = \frac{\Delta(C - C_0)}{\Delta t} \quad (1)$$

$$-k = \frac{\Delta \ln(C/C_0)}{\Delta t} \quad (2)$$

where  $C$  is the measured  $O_2$  or  $NO_3^-$  concentration,  $C_0$  is the initial  $O_2$  or  $NO_3^-$  concentration in recharge [Böhlke, 2002],  $\Delta t$  is the difference between the ground water age of the sample and the age when it entered the relevant reaction zone,  $R$  is the rate (mg/L-year) for zero-order reaction kinetics, and  $k$  (1/a) is the rate constant for first-order reaction kinetics. For  $O_2$  reduction, the water table was assumed to be the top of the reaction zone, and  $C_0$  was assigned the maximum measured  $O_2$  concentration near the water table, which is likely to result in maximum estimates of  $R$  and  $k$ . For denitrification,  $C_0$  is equal to the sum of  $NO_3^-$ -N + excess  $N_2$ -N, where excess  $N_2$ -N is the concentration of  $N_2$  attributed to denitrification. The methods for calculating excess  $N_2$ -N and initial  $NO_3^-$  are presented in Text S1.

[12] In thin reaction zones where denitrification appeared to occur rapidly, denitrification rates were estimated by comparing analytical solutions to the one-dimensional advection-dispersion equation with decay to vertical  $NO_3^-$  concentration profiles using the following models:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - R \quad (3)$$

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - kC \quad (4)$$

where  $D$  is the coefficient of hydrodynamic dispersion,  $z$  is distance along the flow path, and  $v$  is ground water interstitial velocity in the denitrifying zone. Average ground water velocities in the denitrifying zones ranged from 0.7 (Tampa) to 1.6 m/a (York) and were obtained from the ground water flow models. Details of our 1-D modeling approach and the analytical solutions are presented in Text S1.

## 4. Results and Discussion

[13] The quality of water produced by PSW is complexly linked to the quality of water recharging and the effect of transport. By assessing relations between the quality of water recharging the aquifers ( $NO_3^-$  sources), the effect of transport through the hydrologic systems on  $NO_3^-$  ( $NO_3^-$  transport), and the quality of water produced by the PSW ( $NO_3^-$  receptors) in differing hydrogeological settings,

insight can be gained into the factors controlling the occurrence of  $NO_3^-$  in water supplies.

### 4.1. Nitrate Sources

[14] Areas contributing recharge to the four PSW included a variety of land use settings such as agricultural, urban, and forest lands (Table 1). Thus,  $NO_3^-$  in water produced by the PSW could have been derived from numerous sources associated with those land uses. Nitrate under different land uses was characterized by its initial concentration and N isotope composition at the time of recharge (Figure 3), as well as its corresponding apparent ground water age (Figure 4). The  $NO_3^-$  source characterization is based on monitoring well samples. Samples from the PSW also are plotted in Figure 3, but they are discussed in the section "Nitrate Receptors."

#### 4.1.1. Agricultural Sources of Nitrate

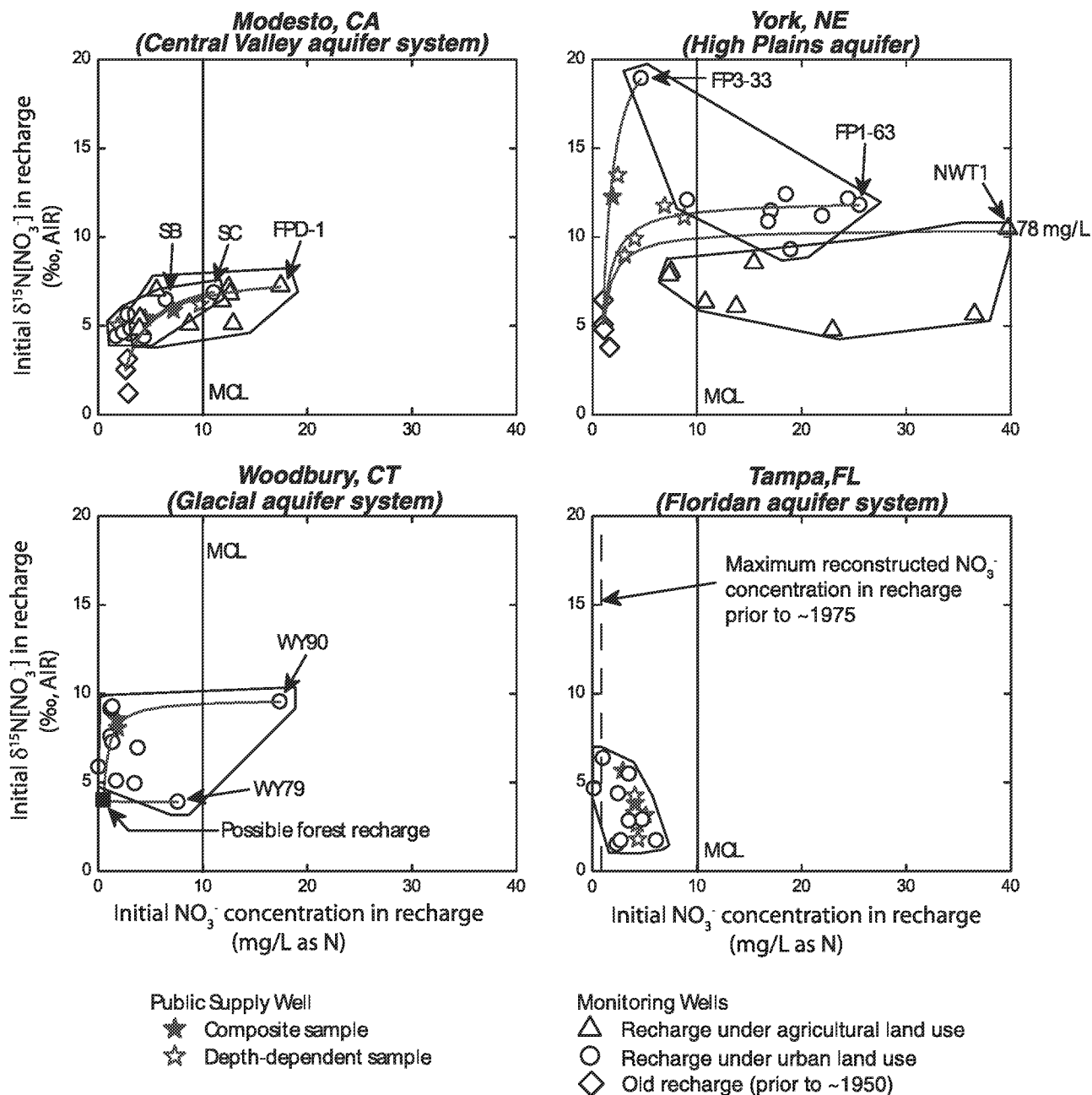
[15] Areas contributing recharge to the PSW in Modesto and York were the only ones to contain >5% agricultural lands (Table 1), so those were the only sites to have monitoring wells installed in agricultural areas. Median  $NO_3^-$  concentrations in agricultural recharge in Modesto and York were 12.5 and 15.7 mg/L as N, respectively (Table 2 and Figure 3). Median values of  $\delta^{15}N[NO_3^-]$  in agricultural recharge in Modesto and York were 6.4 and 7.1‰, respectively.

[16] The values of  $\delta^{15}N[NO_3^-]$  in agricultural recharge from both study areas could be consistent with  $NO_3^-$  derived from natural soil N or from fertilizer N that has undergone varying amounts of isotopic fractionation in the soil zone prior to recharge [Kreitler, 1979; Gormly and Spalding, 1979]. Fertilizer was the primary source of N applied to agricultural fields in both study areas. Furthermore,  $NO_3^-$  concentrations and  $\delta^{15}N$  values in recent agricultural recharge were distinctly different from those in old recharge (>50 years) that may represent natural conditions (Figure 3). Thus, fertilizer is considered to be the most likely source of large  $NO_3^-$  concentrations in agricultural recharge in both study areas. The sample of York agricultural recharge with the largest  $NO_3^-$  concentration (well NWT1 in Figure 3), however, had a  $\delta^{15}N$  value of 10.4‰, more characteristic of an animal waste source, and was located downgradient from a former manure-spreading area, so animal waste sources may be locally important in that agricultural area.

[17] On average, the dated ground water records indicate that  $NO_3^-$  concentrations in agricultural recharge at Modesto and York increased by about 11.5 and 14.5 mg/L as N, respectively, from before 1950 to 1995 (Figure 4). The increases in  $NO_3^-$  concentrations could be related to the temporal increase in farm fertilizer applications over the past ~50 years if the equivalent of about 40 to 50% of the applied N was nitrified and leached to the water table (Figure 4). Similar temporal trends and leaching fractions have been reported for agricultural recharge in other areas in the United States [Hallberg, 1986; Böhlke and Denver, 1995; Böhlke et al., 2002, 2007; Howarth et al., 2002; McMahon et al., 2006].

#### 4.1.2. Urban Sources of Nitrate

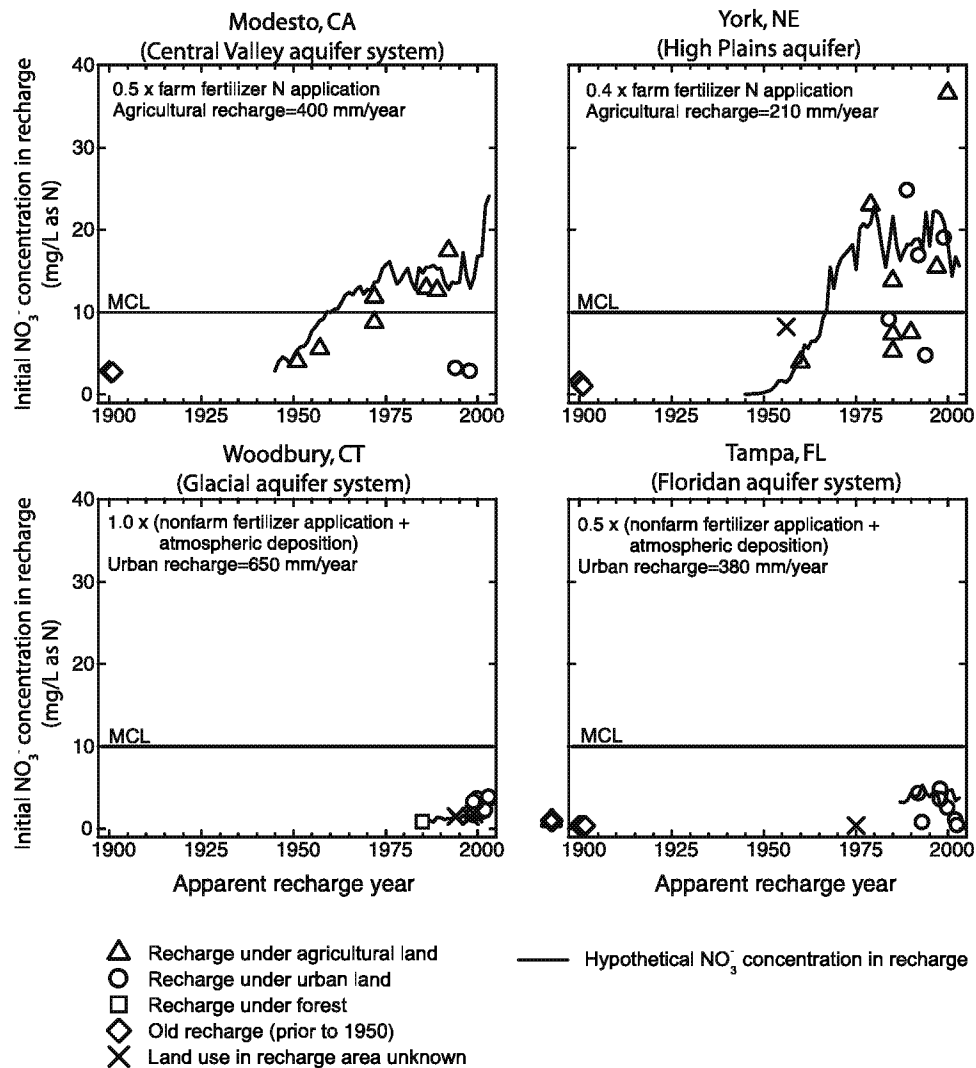
[18] Median  $NO_3^-$  concentrations in urban recharge at the four sites ranged from 1.14 mg/L as N at Woodbury to 22.5 mg/L as N at York (Table 2 and Figure 3). Excluding the York site,  $NO_3^-$  concentrations in urban recharge were



**Figure 3.** Initial nitrate concentrations and nitrogen isotope compositions in recharge to monitoring wells, by land use. Also shown are initial nitrate concentrations and nitrogen isotope compositions for public supply well (PSW) composite and depth-dependent samples. Where necessary, initial concentration and isotope compositions were obtained by correcting measured values for denitrification effects produced in the aquifer (see Text S1 for details). Mixing lines show some possible source contributions to nitrate in PSW samples. The forest end member for mixing in Woodbury is based on data from Grady and Mullaney [1998] and Mayer *et al.* [2002].

much smaller than the concentrations in agricultural recharge. This finding is consistent with a national survey of  $\text{NO}_3^-$  concentrations in shallow ground water in the United States that reported a larger median  $\text{NO}_3^-$  concentration under agricultural land than urban land [Nolan and Stoner, 2000]. Median values of  $\delta^{15}\text{N}[\text{NO}_3^-]$  in urban recharge ranged from 2.9‰ at Tampa to 11.8‰ at York.

[19] The  $\delta^{15}\text{N}$  values for York urban recharge generally were larger than 10‰, which could be characteristic of  $\text{NO}_3^-$  derived from animal waste sources [Kreidler, 1975; Heaton, 1986; McMahon and Böhlke, 1996]. The most likely sources of animal waste N at the York urban area are septic systems and perhaps leaking sanitary sewer lines. The  $\delta^{15}\text{N}$  values for  $\text{NO}_3^-$  in urban recharge at Modesto and



**Figure 4.** Initial nitrate concentrations in recharge in relation to recharge year. Where necessary, initial concentrations were obtained by correcting measured concentrations for denitrification effects produced in the aquifer (see Text S1 for details). Apparent groundwater ages were determined using CFC,  $\text{SF}_6$ , and (or)  $^3\text{H}/^3\text{He}$  data. Samples containing  $<0.5$  TU of  $^3\text{H}$  were assigned recharge dates of  $\sim 1900$ ; however, some samples could be older on the basis of particle ages and  $^{14}\text{C}$  data, or slightly younger (but prior to  $\sim 1950$ ). The solid lines represent nitrate concentrations that would occur in agricultural or urban recharge for the indicated recharge rates and fractions of applied nitrogen leached to the water table. Nitrogen applications were based on county-level data from Ruddy *et al.* [2006]. Farm fertilizer applications were normalized to total cropland in the county (U.S. Department of Agriculture, Census of agriculture: National Agricultural Statistics Service, variously dated, [http://www.nass.usda.gov/Census\\_of\\_Agriculture/index.asp](http://www.nass.usda.gov/Census_of_Agriculture/index.asp)). Atmospheric nitrogen deposition is for wet deposition of nitrate and ammonium [Ruddy *et al.*, 2006]. MCL is Maximum Contaminant Level.

Tampa (Figure 3) are more consistent with  $\text{NO}_3^-$  derived from atmospheric deposition, natural soil N, or fertilizer N than with an animal waste source. It is not likely that atmospheric deposition in the absence of substantial evaporative concentration could produce  $\text{NO}_3^-$  concentrations larger than 1 to 2 mg/L as N in any of the study areas based on total inorganic N concentrations in wet deposition from nearby NADP sites (National Atmospheric Deposition Program, 2006, available at <http://nadp.sws.uiuc.edu/sites/ntnmap.asp?>), average values of which ranged from 0.21 to 0.83 mg/L as N. Likewise, the small  $\text{NO}_3^-$  concentrations in old recharge at Modesto and Tampa (Figures 3 and 4)

indicate that soil N probably was not a source for the largest  $\text{NO}_3^-$  concentrations in urban recharge in those two areas. Hence, fertilizer probably was a more important source of large  $\text{NO}_3^-$  concentrations than soil N at Modesto and Tampa. The Modesto urban monitoring well with the largest  $\text{NO}_3^-$  concentrations (well SC in Figure 3), however, was located in an unsewered part of the city. Thus, it is possible that septic leachate also was an important source of large  $\text{NO}_3^-$  concentrations in Modesto urban recharge.

[20] At Woodbury, the  $\delta^{15}\text{N}$  values for  $\text{NO}_3^-$  in urban recharge (Figure 3) indicate that fertilizer, soil, and animal waste all could have been possible sources of  $\text{NO}_3^-$  in that

area. Nitrate concentrations in forest recharge in Connecticut and other northeastern States typically are less than 1 mg/L as N (Figure 4) [Grady and Mullaney, 1998; Mayer *et al.*, 2002], so soil N was not a likely source of the large  $\text{NO}_3^-$  concentrations in recharge. The Woodbury sample with the largest  $\text{NO}_3^-$  concentration (well WY90 in Figure 3) was located downgradient from residential septic systems and had a  $\delta^{15}\text{N}$  value (9.5‰) that is consistent with a septic source of N. The Woodbury sample with the second largest  $\text{NO}_3^-$  concentration (well WY79 in Figure 3) had a  $\delta^{15}\text{N}$  value (3.9‰) that is more consistent with a fertilizer source of N.

[21] Nitrate concentrations in urban recharge at Tampa could be from nonfarm fertilizer applications and atmospheric N deposition if the equivalent of ~50% of that N was nitrified and leached to the water table (Figure 4). In the case of Woodbury, the available data indicate that nonfarm fertilizer and atmospheric N deposition could not account for most of the large  $\text{NO}_3^-$  concentrations in urban recharge, even if 100% of that N leached to the water table. This apparent discrepancy probably could be resolved if  $\text{NO}_3^-$  loading from septic systems in that area was accounted for. Septic systems are the primary waste disposal system used in that community.

#### 4.1.3. Nitrate Recharge Fluxes

[22] Sources of  $\text{NO}_3^-$  in the areas contributing recharge to the PSW were diverse and appear to have included farm and nonfarm fertilizers, septic leachate or leaking sewer lines, manure spreading, atmospheric deposition, and natural soil N. Nitrate fluxes to the water table depended on the strength of those sources and recharge rates. Nitrate recharge fluxes were calculated using  $\text{NO}_3^-$  concentrations in recharge and water balance estimates of recharge (Table 2). Variability in the flux estimates probably represents spatial and temporal variations in  $\text{NO}_3^-$  inputs and recharge rates (Figures 3 and 4). Median  $\text{NO}_3^-$  recharge fluxes under agricultural land (33 to 69 kg N/ha-year) were larger than those under urban land (7.4 to 16 kg N/ha-year) (Table 2). The agricultural  $\text{NO}_3^-$  fluxes were similar to or larger than agricultural  $\text{NO}_3^-$  fluxes determined similarly in Minnesota, Maryland, and Nebraska (32 to 46 kg N/ha-year) [Böhlke and Denver, 1995; Böhlke *et al.*, 2002, 2007], and the largest urban  $\text{NO}_3^-$  flux was similar to the  $\text{NO}_3^-$  flux under Nottingham, England (21 kg N/ha-year) [Wakida and Lerner, 2005]. Although the  $\text{NO}_3^-$  recharge fluxes reported here are only approximations, they imply that agricultural sources produced larger  $\text{NO}_3^-$  loads to ground water than urban sources. From the standpoint of minimizing  $\text{NO}_3^-$  contamination in PSW, these findings indicate that it would be beneficial to reduce PSW capture zone areas in agricultural regions. In urban settings, consideration should be given to understanding the distribution of septic systems and leaking sewer lines in PSW capture zones.

## 4.2. Nitrate Transport

### 4.2.1. Terminal Electron-Accepting Processes

[23] Concentrations of  $\text{O}_2$  in water from the aquifers at Modesto and Woodbury were mostly  $\geq 0.5$  mg/L, whereas waters containing  $< 0.5$  mg/L  $\text{O}_2$  were more widespread in the aquifers at York and Tampa. For  $\text{NO}_3^-$  transport, the transition zone between oxic and anoxic conditions in an aquifer is important because that is generally where microbially mediated nitrate reduction begins [Korom, 1992].

Chapelle *et al.* [1995] proposed an  $\text{O}_2$  concentration of 0.5 mg/L as the threshold at which  $\text{NO}_3^-$  reduction begins, but onset of  $\text{NO}_3^-$  reduction has been reported in aquifers where field measurements of  $\text{O}_2$  concentrations were of the order of 1–2 mg/L [Böhlke *et al.*, 2002; McMahon *et al.*, 2004; Böhlke *et al.*, 2007]. In this study, oxic waters are operationally defined by  $\text{O}_2 \geq 0.5$  mg/L. Data from all the sites show that excess  $\text{N}_2$  concentrations were significantly smaller in samples containing  $\geq 0.5$  mg/L  $\text{O}_2$  (average of 0.6 mg N/L) than in samples containing  $< 0.5$  mg/L  $\text{O}_2$  (average of 1.7 mg N/L) ( $p = 0.002$ , Mann-Whitney test).

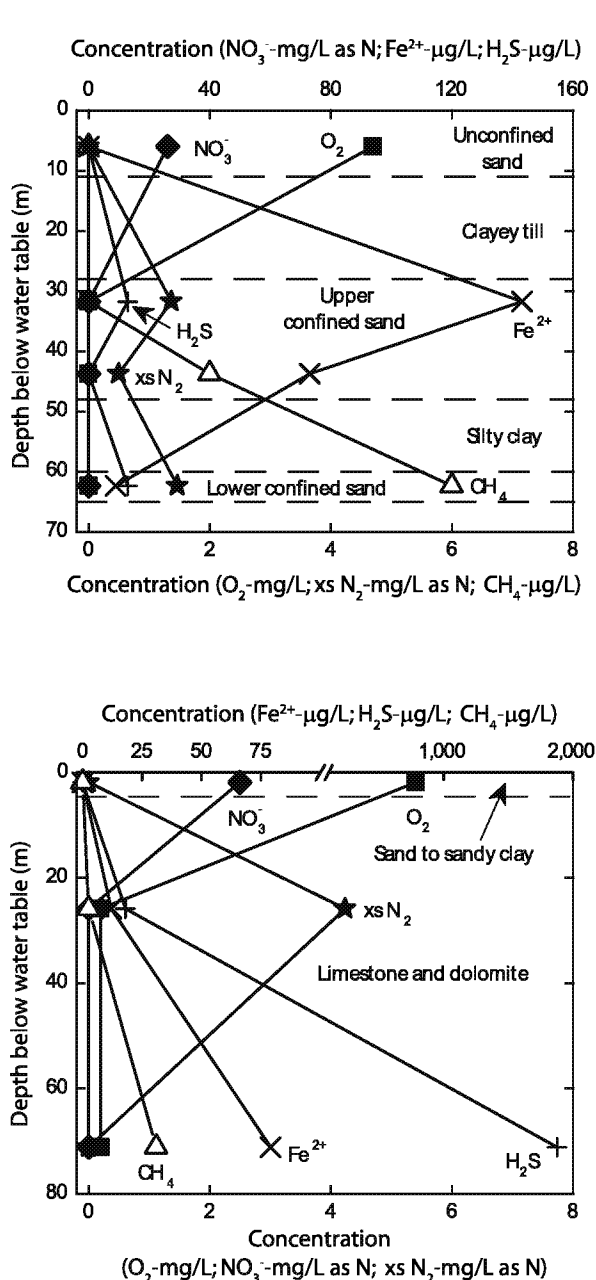
[24] The selected PSW at Modesto and Woodbury were screened in oxic zones (Figure 2). The general absence of anoxic conditions in the aquifers at Modesto and Woodbury indicates that  $\text{NO}_3^-$  was likely to persist as it was transported along flow paths from recharge areas to those PSW. At York, the transition zone between oxic and anoxic conditions was located in the clayey till confining unit between the base of the unconfined sand and the top of the upper confined sand (Figure 2). At Tampa, the transition zone was located near the top of the Floridan aquifer system, just a few meters below the water table, and probably is related to the downward fining of sediment texture in the surficial deposits. The selected PSW at York and Tampa were screened below the redox transition zones, indicating that  $\text{NO}_3^-$  was likely to be at least partially reduced as it was transported along flow paths from recharge areas to those PSW.

[25] Vertical profiles of redox-sensitive species at York and Tampa show that concentrations of excess  $\text{N}_2$  increased with depth below the water table as concentrations of  $\text{O}_2$  decreased (Figure 5), indicating that reduction of  $\text{NO}_3^-$  did occur along flow paths leading to the producing intervals in which those PSW were screened. Median denitrification reaction progress in the PWS production zones ranged from 0 to 94 percent (Figure 6), with the greatest progress occurring in the aquifers at York and Tampa. The presence of elevated  $\text{Fe}^{2+}$ ,  $\text{H}_2\text{S}$ , and  $\text{CH}_4$  concentrations indicate that anoxic zones at York and Tampa progressed beyond denitrification to Fe(III) reduction,  $\text{SO}_4^{2-}$  reduction, and methanogenesis (Figure 5).

### 4.2.2. Isotopic Evidence for Denitrification

[26] The N and O isotopes of  $\text{NO}_3^-$  in oxic water samples, which included almost all of the samples from Modesto and Woodbury, exhibited little or no evidence of fractionation (Figure 7). At York and Tampa, N and O isotopes in anoxic water are positively correlated and resemble fractionation trends that have been reported for denitrification [e.g., Böttcher *et al.*, 1990; Aravena and Robertson, 1998; Mengis *et al.*, 1999; Fukada *et al.*, 2003; Sigman *et al.*, 2005]. Least squares regressions yield apparent fractionation trends ( $\Delta\delta^{18}\text{O}/\Delta\delta^{15}\text{N}$ ) of approximately 0.5 for York (near the low end of the reported range for denitrification) and 0.83 for Tampa (near the high end of the reported range). Scatter in these trends may be attributed in part to variations in the initial values of  $\delta^{18}\text{O}$  and  $\delta^{15}\text{N}$  for individual samples.

[27] The N isotope compositions of residual  $\text{NO}_3^-$  and excess  $\text{N}_2$  plotted relative to denitrification reaction progress are qualitatively consistent with fractionations expected for denitrification (Figure 6). The apparent enrichment factors ( $\epsilon$ ) that would be consistent with the data are approximately –10 to –2‰. These apparent  $\epsilon$  are variable



**Figure 5.** Concentration profiles for redox-sensitive species in ground water in relation to depth below the water table at selected monitoring well nests at (a) York, Nebraska, and (b) Tampa, Florida; xs  $\text{N}_2$  refers to excess  $\text{N}_2$  from denitrification.

and generally smaller than those determined experimentally for denitrification (about  $-30$  to  $-20\text{‰}$ ) but within the range of possibilities in heterogeneous ground water systems [e.g., Mariotti *et al.*, 1988].

#### 4.2.3. Rates of Oxygen Reduction and Denitrification in Unconfined Sands

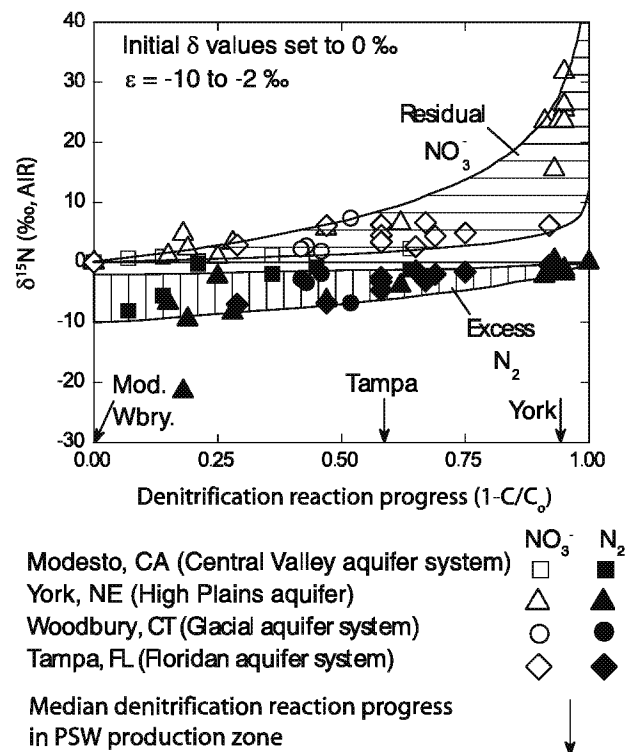
[28] The presence of relatively large  $\text{O}_2$  concentrations in water from the Turlock Lake and Riverbank Formations in Modesto, unconfined sand in York, glacial drift in Woodbury, and surficial sand in Tampa (Figure 2) indicate that those coarse sediments could represent thick reaction zones

where  $\text{O}_2$  reduction and denitrification occur gradually. Thus, equations (1) and (2) were used to describe the reaction kinetics. Results for both kinetic models (zero-order and first-order) are presented in Table 3 for comparison with other data, although it is uncertain whether either of these is the correct model because of sparse data in the reaction zone. The following comparative discussion focuses on the first-order rate constants,  $k$ .

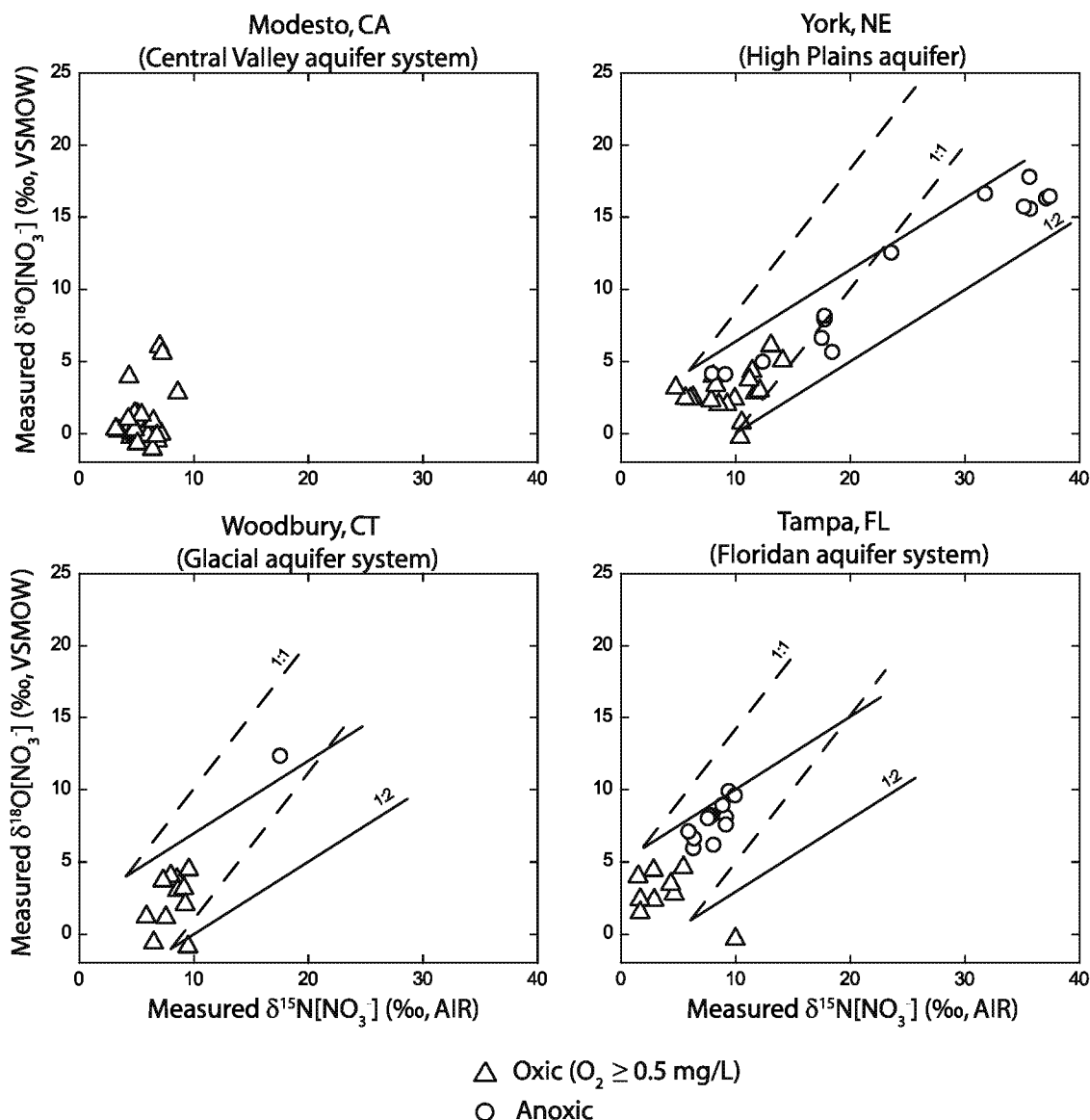
[29] Rate constants for  $\text{O}_2$  reduction at York, Woodbury, and Tampa were roughly similar and ranged from 0.02 to  $1/a$  (Figure 8). The values at those sites are similar to  $\text{O}_2$  reduction rate constants reported for fluvial (0.1/a) and glaciofluvial (0.13/a) sands in Nebraska and Minnesota, respectively [Böhlke *et al.*, 2002, 2007]. The largest rate constant at Modesto, 0.03/a, was at the low end of values at the other sites. Rate constants for  $\text{O}_2$  reduction in the study areas have important implications with respect to lag times for the start of denitrification.

[30] Excess  $\text{N}_2$  was present in small concentrations (0.3 to 1.8 mg/L as N) in the aquifer at Modesto and the concentrations systematically increased with depth at multiple well nests, indicating that small amounts of denitrification occurred in the aquifer even though it was generally oxic. All of the samples with detectable excess  $\text{N}_2$  had apparent ground water ages  $\geq 40$  years (Figure 8), indicating a lag time of possibly several decades before the onset of

**B.**



**Figure 6.** Nitrogen isotope compositions of residual nitrate and excess  $\text{N}_2$  in relation to denitrification reaction progress. Initial (undenitrified)  $\delta$  values for nitrate were set to 0‰ to facilitate comparisons between aquifers. Hatched areas represent Rayleigh-type fractionation trends with enrichment factors ranging from  $-10$  to  $-2\text{‰}$ . The method for calculating denitrification reaction progress is presented in Text S1.



**Figure 7.** Measured nitrogen and oxygen isotope compositions of nitrate in ground water. The lines indicate the range of fractionation trends reported for nitrate reduction. ( $\Delta\delta^{18}\text{O}/\Delta\delta^{15}\text{N} \approx 0.5$  to 1.0).

denitrification. Such a long lag time may be related to the relatively small rate constants for  $\text{O}_2$  reduction at that site. Much longer lag times prior to denitrification have been reported elsewhere (e.g.,  $\sim 3,000$ – $10,000$  years [Vogel *et al.*, 1981; McMahon *et al.*, 2004]). Values of  $\ln(C/C_0)$  plotted in relation to ground water age correspond to a maximum  $k$  of 0.02/a at Modesto (Figure 8), assuming a lag time of  $\sim 30$  years after recharge before the onset of denitrification. This lag time corresponds to the oldest sampled water at Modesto that did not contain detectable excess  $\text{N}_2$  (Figure 8). This analysis is limited by the fact that the most denitrified samples, which were the deepest samples in their respective well nests, were too old to be dated with the available tracers and so were omitted from the analysis. One possible explanation for denitrification in this oxic flow system is that it occurred in association with abundant sand/clay contacts in the aquifer. Clay and silt

comprise about 50% of the sediments in this aquifer. Despite the apparent occurrence of denitrification in the Central Valley aquifer, its rate was sufficiently slow to allow 95%, on average, of the  $\text{NO}_3^-$  originally present in modern recharge to persist along the sampled flow paths.

[31] For the most part, excess  $\text{N}_2$  was not detected in the unconfined, sandy sediments at the other three sites (Figure 8), indicating a general lack of denitrification in those settings even though  $\text{O}_2$  reduction rate constants were larger there than at Modesto (Table 3). Larger  $\text{O}_2$  reduction rate constants presumably would produce anoxic conditions more quickly at those sites than at Modesto. At Woodbury and Tampa, the apparent lack of denitrification could result from the short residence time (roughly  $<10$  years) of water in those sandy sediments (Figure 8). Ground water residence times in the unconfined sand in York were as long as

**Table 3.** Apparent Rates and Rate Constants for O<sub>2</sub> Reduction and Denitrification<sup>a</sup>

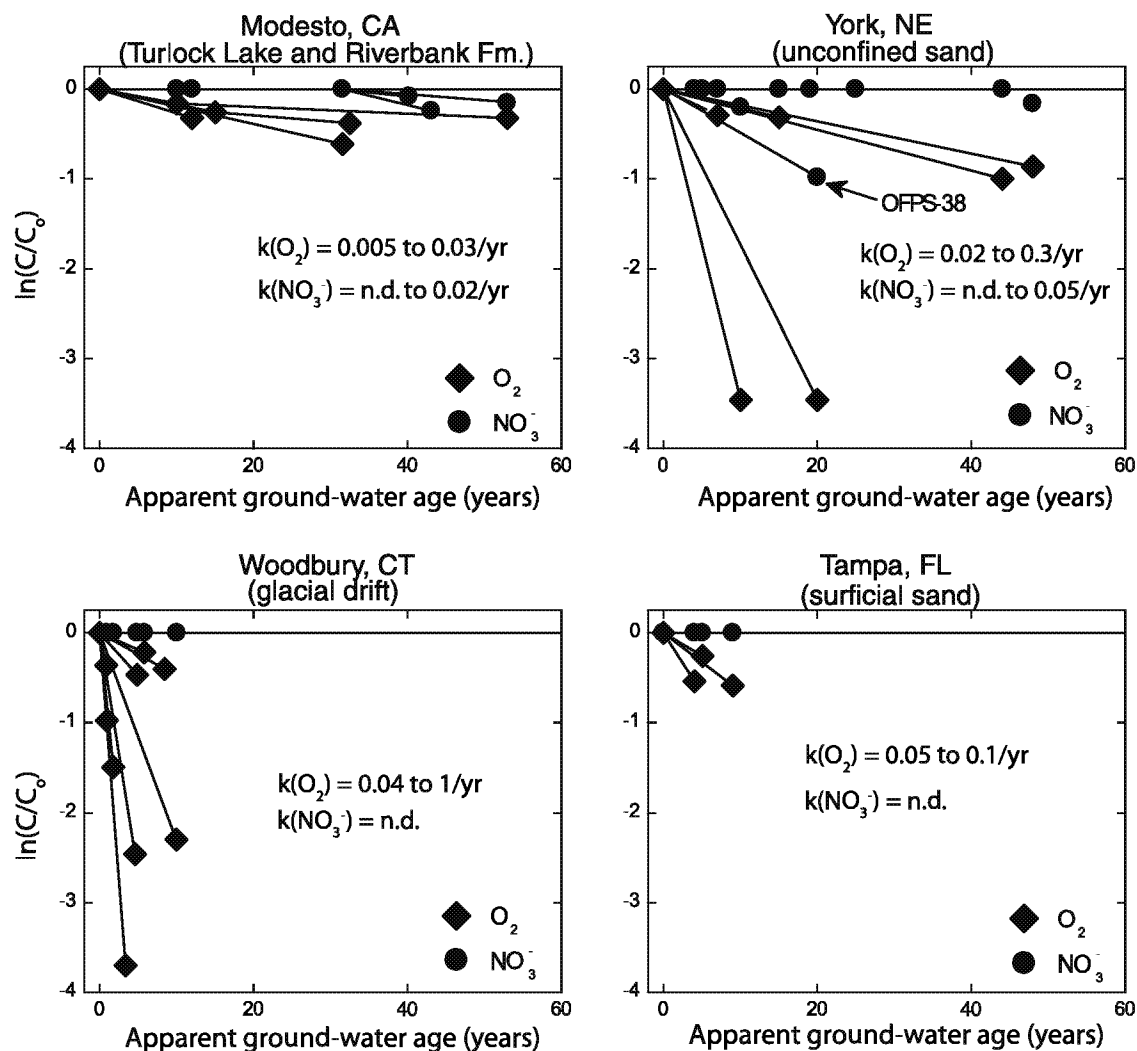
Variable	York, Nebraska							Tampa, Florida		
	Modesto, California	Unconfined Sand	Clayey Till	Upper Confined Sand	Silty Clay	Carlie Shale	Woodbury, Connecticut	Unconfined Sand	Sandy Clay/Clayey Sand	Carbonates
O <sub>2</sub> reduction rate, <i>R</i> (mg/L-year)	0.03 to 0.2	0.08 to 0.6	—	— <sup>b</sup>	— <sup>b</sup>	—	0.3 to 6	0.2 to 0.4	—	— <sup>b</sup>
O <sub>2</sub> reduction rate constant, <i>k</i> (1/a)	0.005 to 0.03	0.02 to 0.3	—	— <sup>b</sup>	— <sup>b</sup>	—	0.04 to 1	0.05 to 0.1	—	— <sup>b</sup>
Denitrification lag time <sup>c</sup> (years)	30	40	—	—	—	—	Not applicable	5 to 10	—	—
Denitrification rate, <i>R</i> (mg-N/L-year)	n.d. <sup>d</sup> to 0.07	n.d. <sup>d</sup> to 6	0.2 to 14	— <sup>b</sup>	— <sup>b</sup>	—	n.d. <sup>d</sup>	n.d. <sup>d</sup>	0.1 to 0.8	— <sup>b</sup>
Denitrification rate constant, <i>k</i> (1/a)	n.d. <sup>d</sup> to 0.02	n.d. <sup>d</sup> to 0.05	0.1 to 6	— <sup>b</sup>	— <sup>b</sup>	—	n.d. <sup>d</sup>	n.d. <sup>d</sup>	0.1 to 0.5	— <sup>b</sup>

<sup>a</sup>Also listed are apparent denitrification lag times (—, not measured; n.d., no denitrification detected).

<sup>b</sup>Most sampled intervals in this hydrogeologic unit did not contain detectable NO<sub>3</sub><sup>−</sup> (none contained detectable O<sub>2</sub>).

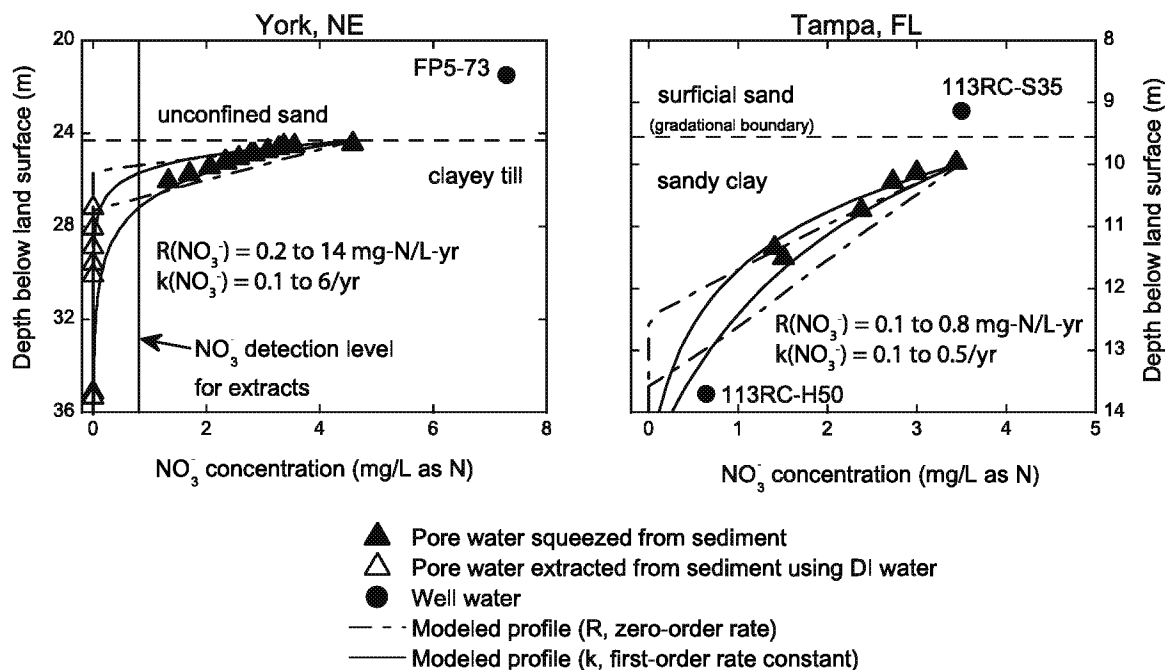
<sup>c</sup>Lag time is the time of travel of a groundwater particle through the O<sub>2</sub> reduction zone prior to the onset of denitrification; lag time could be shorter than indicated where localized conditions support denitrification.

<sup>d</sup>O<sub>2</sub> concentration >3 mg/L in sampled interval.



**Figure 8.** Range of apparent rate constants (*k*), assuming first-order kinetics (equation (2) in text), for oxygen reduction and denitrification in aquifer sediments. Samples are from monitoring wells; n.d. indicates “no denitrification” detected in samples on the basis of excess N<sub>2</sub> concentrations.





**Figure 9.** Range of apparent rates ( $R$ ) and rate constants ( $k$ ), assuming zero- and first-order kinetics, respectively, for denitrification in confining units in the High Plains and Floridan aquifer systems. Core was collected from site FP5 near York and from site 113RC near Tampa. Values of  $R$  and  $k$  were estimated by comparing analytical solutions to the one-dimensional advection-dispersion equation with degradation (equations (3) and (4) in text) to the vertical nitrate concentration profiles.

those at Modesto but systematic increases in excess  $\text{N}_2$  with depth were not detected at York like they were at Modesto. The reason for this difference between the two sites is not clear, but it could be related to the smaller clay content in the sediment at York compared to Modesto if the clays contain more organic carbon to support denitrification than sands [McMahon *et al.*, 1999].

[32] Denitrification could be locally important in the unconfined sands at York, Woodbury, and Tampa, in places where lag times may be considerably shorter. For example, water from well OFPS-38 at York (Figure 8), which is located in the floodplain of a small stream, contained no detectable  $\text{O}_2$ , 5.7 mg/L of excess  $\text{N}_2\text{-N}$  and elevated concentrations of  $\text{Fe}^{2+}$  and  $\text{CH}_4$ . The apparent age of that water was about 20 years, but denitrification is likely to have begun soon after recharge in the anoxic floodplain sediments.

#### 4.2.4. Rates of Oxygen Reduction and Denitrification in Clay Confining Units

[33] The presence of relatively small  $\text{O}_2$  concentrations in the clayey till at York and sandy clay above limestone at Tampa (Figure 2) indicate that those sediments could represent thin reaction zones where  $\text{O}_2$  reduction and denitrification occur rapidly. Thus, equations (3) and (4) were used to describe the reaction kinetics. At York, the modeled denitrification zone was the top of the clayey till confining unit at well site FP5, which was a transition zone from oxic to anoxic conditions at that site (Figure 2). At Tampa, the modeled zone was the top of the sandy clay at well site 113RC, which was the transition zone from oxic to anoxic conditions at that site (Figure 2).

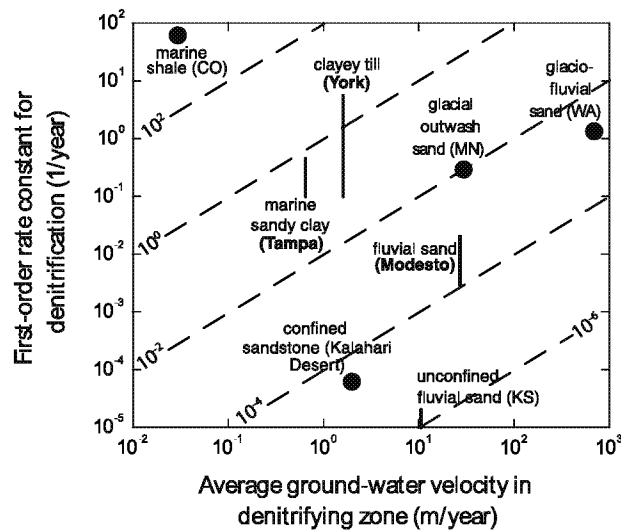
[34] Nitrate concentrations in the sediment pore waters at both sites decreased with depth, implying that denitrification may have occurred (Figure 9). Other evidence for denitrification in those pore waters included fractionation

of N and O isotopes in residual  $\text{NO}_3^-$ , accumulation of excess  $\text{N}_2$ , and (or) a decrease in  $\text{NO}_3^-/\text{Cl}^-$  ratios with depth. The decrease in  $\text{NO}_3^-$  concentrations with depth at both sites appears to be better represented by the models with first-order reaction kinetics than the ones with zero-order kinetics (Figure 9). In these instances,  $k$  ranged from 0.1 to 6/a at York and from 0.1 to 0.5/a at Tampa. These rate constants are considerably larger than those from the Modesto and Woodbury sandy sediments (Table 3), which lack laterally extensive confining layers. Thus, the confining layers at York and Tampa are considered to be more effective reactive barriers to  $\text{NO}_3^-$  transport than isolated denitrifying zones that might be present in the unconfined sands at Modesto and Woodbury. Furthermore, Fe(III)-reducing,  $\text{SO}_4^{2-}$ -reducing, and methanogenic conditions deeper in the aquifers at York and Tampa (Figure 5) imply that  $\text{NO}_3^-$  transported below the confining layers would not persist under natural conditions.

[35] The spatial distribution of denitrifying zones in the aquifers at York and Tampa differed markedly from the spatial distribution of denitrifying zones in a shallow sand aquifer at Cape Cod, Massachusetts. Bowen *et al.* [2007] reported that denitrification was focused near the shallow water table in that aquifer because the source of electrons for denitrification was DOC leached from the soil zone. At York and Tampa, the electron donors for denitrification had geologic sources deeper in the flow system. Thus, there was a ~5- to 40-year lag time prior to the onset of denitrification as ground water traveled through the  $\text{O}_2$  reduction zone near the water table.

#### 4.2.5. Nitrate Attenuation Efficiency

[36] The concentration of  $\text{NO}_3^-$  eventually entering PSW screens depends in part on denitrification rates and ground water velocities along all flow paths leading to the PSW.



**Figure 10.** Nitrate attenuation efficiencies (1/m) represented by the ratio of first-order denitrification rate constant ( $k$ , 1/a) to average groundwater velocity ( $v$ , m/a). The attenuation efficiencies essentially represent fractional nitrate losses from denitrification per unit distance of transport within active denitrification zones. Data are from this study and from Vogel *et al.* [1981], McMahon *et al.* [1999, 2004], Tesoriero *et al.* [2000], and Böhlke [2002].

Nitrate concentrations should be relatively small where denitrification rates outpace velocities and relatively large where velocities outpace rates. Denitrification rate constants and average ground water velocities in denitrifying zones were used as a framework for comparing  $\text{NO}_3^-$  attenuation efficiency, defined as  $k/v$ , in the aquifers (Figure 10). The attenuation efficiencies essentially represent fractional  $\text{NO}_3^-$  losses from denitrification per unit distance of transport. General patterns in the  $\text{NO}_3^-$  attenuation efficiencies appear to be related to sedimentary depositional environment; attenuation efficiencies increased as follows: fluvial sand aquifers < glacial sand aquifers < glacial/marine clay < marine shale. Various uncertainties are associated with the  $\text{NO}_3^-$  attenuation efficiencies defined here, such as representative sampling of the reactive zone, ability to date ground water at appropriate timescales, and  $\text{NO}_3^-$  transport limitations on denitrification. Nevertheless, this approach could provide a useful framework for comparing aquifer susceptibility to  $\text{NO}_3^-$  contamination.

### 4.3. Nitrate Receptors

#### 4.3.1. Nitrate Concentrations in Public Supply Wells

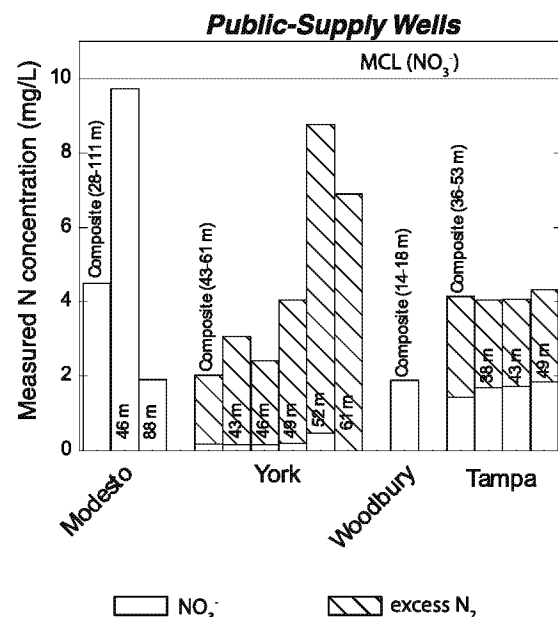
[37] Measured  $\text{NO}_3^-$  concentrations in the PSW composite (wellhead) and depth-dependent samples ranged from <0.06 to 9.7 mg/L as N (Figure 11). The largest measured  $\text{NO}_3^-$  concentrations (4.4 to 9.7 mg/L as N) occurred in samples from the Modesto PSW, and the smallest concentrations (<0.06 to 0.47 mg/L as N) occurred in samples from the York PSW. Denitrification apparently did not reduce any  $\text{NO}_3^-$  to  $\text{N}_2$  in water captured by the Modesto and Woodbury PSW (Figure 11), whereas it reduced about 60 to 95%, on average, of the  $\text{NO}_3^-$  in water captured by the Tampa and York PSW, respectively.

[38] Reconstructed initial  $\text{NO}_3^-$  concentrations (indicated by the sum of  $\text{NO}_3^-$ -N + excess  $\text{N}_2$ -N concentrations in

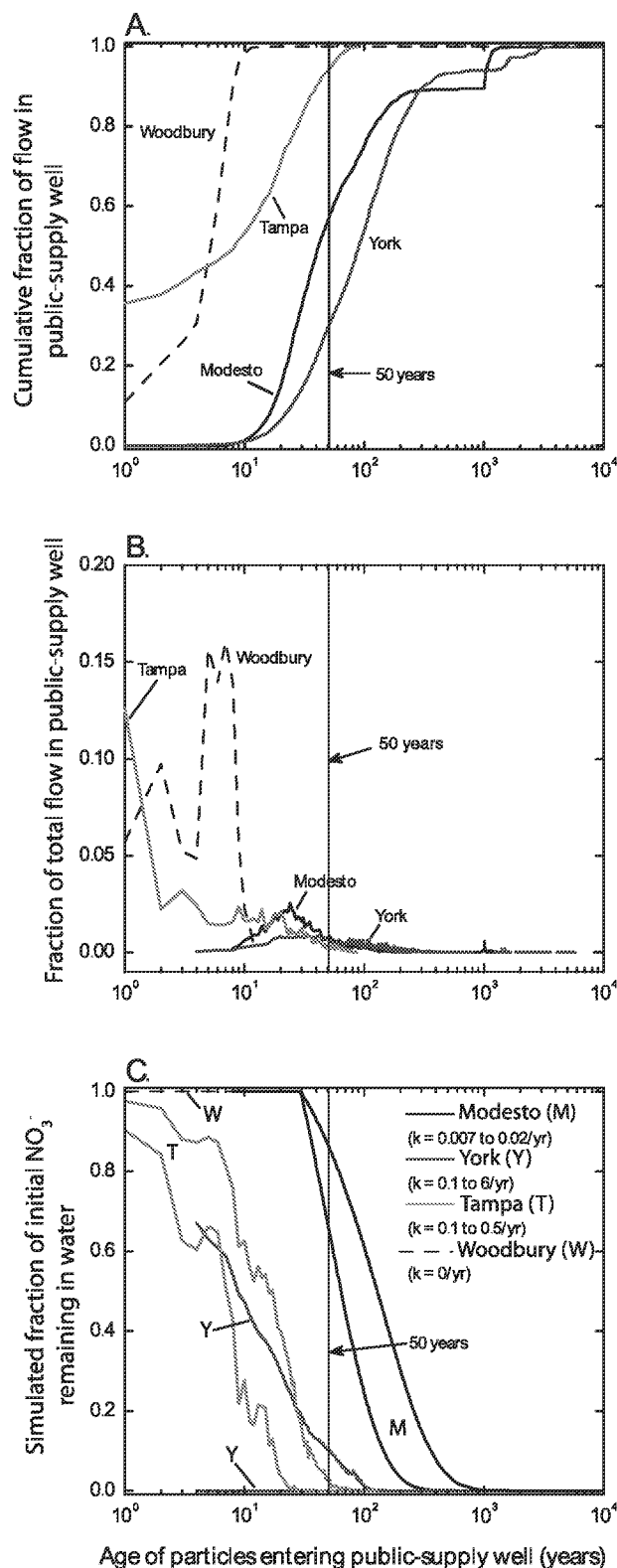
Figure 11) exhibited substantial variability with depth in the well screens at Modesto and York. At Tampa, the lower third of the well intercepted a high-permeability conduit [Katz *et al.*, 2007] (represented by the 49-m sample) that may have dominated the chemistry of shallower depth-dependent samples. The short screen in the Woodbury PSW (Figure 2) precluded the collection of depth-dependent samples from that well. Initial  $\text{NO}_3^-$  concentrations in samples from the Modesto and York PSW were not consistently larger than those from the Tampa and Woodbury PSW even though  $\text{NO}_3^-$  fluxes to the water table at Modesto and York were larger than those at the other two sites (Table 2), indicating that water entering the PSW probably contained  $\text{NO}_3^-$  from multiple sources and (or) water of multiple ages.

#### 4.3.2. Mixing in Public Supply Wells

[39] According to the MODPATH analysis, water from most of the PSW consisted of a broad distribution of ages—spanning <20 years at Woodbury to >1000 years at Modesto and York (Figure 12a). Contributions of <50-year-old water to flow in the PSW ranged from about 30% at York to 100% at Woodbury (Figure 12a). The <50-year-old fraction of these distributions is important because it contained the largest  $\text{NO}_3^-$  concentrations (Figure 4) and the >50-year-old fraction is important because it could dilute anthropogenic  $\text{NO}_3^-$  concentrations when waters of different age mixed in the PSW. Young ground water also is important because it could indicate limited denitrification potential in flow systems with long denitrification lag times (Table 3). For example, the single largest annual age fraction of water entering the Modesto PSW was about



**Figure 11.** Concentrations of nitrate and excess  $\text{N}_2$  in composite samples from the PSW wellhead and from depth-dependent samples in the PSW. The composite samples represent the entire screened interval, shown in parentheses. The depth-dependent samples are from specific depths, shown in parentheses, in the PSW well screen. MCL is Maximum Contaminant Level.



25 years old (Figure 12b), which is shorter than the 30-year denitrification lag time at that site (Table 3). Thus, NO<sub>3</sub><sup>-</sup> in that young fraction of water is less likely to have been denitrified compared to NO<sub>3</sub><sup>-</sup> in older fractions of water.

[40] All of the PSW samples contained some anthropogenic NO<sub>3</sub><sup>-</sup> on the basis of mixing calculations in which initial NO<sub>3</sub><sup>-</sup> concentrations and isotope compositions in recharge were used to define possible mixing end members (Figure 3). Most of the Modesto PSW samples plot along mixing lines defined by old recharge and contaminated modern urban (well SC) or agricultural (well FPD-1) recharge. Twenty to 55% of the water in the composite samples (and 55 to 85% of the NO<sub>3</sub><sup>-</sup>) may have been derived from urban sources such as septic leachate (as represented by SC). Alternatively, 15 to 30% of the water in the composite samples (50 to 75% of the NO<sub>3</sub><sup>-</sup>) may have been derived from agricultural sources such as irrigation return flow containing fertilizer N (as represented by FPD-1). It also is possible that the composite samples contained NO<sub>3</sub><sup>-</sup> from both anthropogenic sources. The remaining NO<sub>3</sub><sup>-</sup> presumably was from old, possibly natural, sources. These fractions of modern anthropogenic recharge in the Modesto PSW samples are roughly consistent with the simulated fraction of <50-year-old water entering the Modesto PSW (Figure 12a).

[41] Old recharge (>50 years) apparently was not present in the aquifer at Woodbury, but recharge through forest soils in New England generally contains small NO<sub>3</sub><sup>-</sup> concentrations (<1 mg/L as N) [Grady and Mullaney, 1998; Mayer *et al.*, 2002]. The Woodbury PSW samples plot along a mixing line defined by hypothetical forest recharge and contaminated urban recharge (WY90) (Figure 3), indicating that only about 10% of the water in those composite samples (but 75% of the NO<sub>3</sub><sup>-</sup>) may have been derived from urban sources such as septic leachate (as represented by WY90). The composite samples do not plot near the mixing line defined by well WY79 (Figure 3), which may contain NO<sub>3</sub><sup>-</sup> from a fertilizer source.

[42] The York PSW samples generally plot along mixing lines defined by old recharge and two urban end members (wells FP1-63 and FP3-33 in Figure 3), such that 25% of the water in the composite sample (60% of the NO<sub>3</sub><sup>-</sup>) may have been derived from urban sources such as septic leachate (as defined by FP1-63). These results are consistent with particle-tracking simulations for the York PSW, which showed that about 30% of the water entering the well was <50 years old (Figure 12a).

[43] At Tampa, the most likely recharge end members containing small NO<sub>3</sub><sup>-</sup> concentrations were old recharge and

**Figure 12.** (a and b) Fractions of flow in the PSW. (c) Simulated fractions of initial nitrate remaining in water captured by the PSW as a function of the simulated age of particles entering the PSW. In Figure 12c,  $k$  refers to the first-order denitrification rate constant within a zone of active denitrification (ranges of values indicate uncertainty of the estimations). Along each flow path, the value of  $k$  was zero before the onset of denitrification. It was assumed that the rate constants measured in the overlying confining layers were representative of the rate constants in the underlying sediments.

modern recharge from wetlands/rivers. The  $^{15}\text{N}$  composition of those types of waters could not be analyzed because of their small  $\text{NO}_3^-$  concentrations and (or) small amounts of excess  $\text{N}_2$ . Nevertheless, the initial  $\text{NO}_3^-$  concentrations and isotopic compositions in Tampa PSW samples indicate that a substantial fraction of  $\text{NO}_3^-$  in those samples probably was derived from urban sources like fertilizer (Figure 3).

#### 4.3.3. Short-Circuiting Pathways at York and Tampa

[44] The York PSW produced waters that were recharged with 2.0 to 8.8 mg/L  $\text{NO}_3^-$  as N (Figure 11), whereas four of seven monitoring wells in that interval produced water that was recharged with  $<2$  mg/L  $\text{NO}_3^-$  as N and 6 of 8 wells contained  $<1$  TU  $^3\text{H}$ . Such data indicate that much of the confined part of the High Plains aquifer may have contained old water that was recharged with substantially less  $\text{NO}_3^-$  than water captured by the PSW. The youngest particle age for water moving downward through the confining unit by advection was about 60 years, further suggesting that under natural flow conditions the confined aquifer should not have contained modern, high- $\text{NO}_3^-$  water. Clark *et al.* [2007] proposed that the occurrence of modern recharge in the confined aquifer resulted from downward leakage of water through wells screened in multiple geologic layers and through the annular space of well bores that penetrated the confining layer, essentially bypassing the confining unit. If that interpretation is correct, the leakage probably did not occur at the PSW itself because the deepest depth-dependent samples contained the largest initial  $\text{NO}_3^-$  concentrations (Figure 11). It is more likely that leakage between aquifers occurred at upgradient locations in the PSW capture zone through high-permeability features such as irrigation wells. Even though  $\text{NO}_3^-$ -rich water may have either partially or fully bypassed the denitrifying zone in the confining unit, about 95% of the initial  $\text{NO}_3^-$  in water captured by the PSW was removed by denitrification (Figure 11). Thus, it appears that denitrification must have occurred within the confined sand where highly reducing conditions naturally existed (Figure 5a).

[45] As at York, waters from the Tampa PSW contained substantially larger initial  $\text{NO}_3^-$  concentrations than several monitoring wells screened in the same producing interval. The PSW samples were recharged with about 4 mg/L  $\text{NO}_3^-$  (Figure 11), whereas 7 of 12 monitoring wells in that interval produced water that was recharged with  $<1$  mg/L  $\text{NO}_3^-$  as N, and 6 of 10 wells contained  $<1$  TU  $^3\text{H}$ . The high-permeability conduit at a depth of about 49 m that was intercepted by the PSW apparently was connected to the surficial aquifer allowing for the rapid movement of modern recharge to the PSW. Katz *et al.* [2007] estimated that 50 to 70% of the water produced from the PSW was from the surficial sand aquifer, rather than the Floridan aquifer, on the basis of chemical mass balance calculations. Those chemical results are consistent with the particle-tracking results (which accounted for conduit flow) that showed the PSW capturing a large fraction of very recent recharge (Figures 12a and 12b). Conduit flow such as this could bypass the denitrifying zone at the top of the Floridan aquifer and also reduce reaction time in the underlying carbonate sediments which were highly reducing (Figure 5b). All of these factors may explain why the PSW contained detectable  $\text{NO}_3^-$  but most monitoring wells in the same interval did not (Figure 3).

#### 4.3.4. Nitrate Age Distributions in Public Supply Wells

[46] Denitrification rates, spatial distribution of redox conditions, and time spent by water in different redox zones strongly influenced the fractions of initial  $\text{NO}_3^-$  remaining in different age fractions of water captured by the PSW. The combined effect of these factors resulted in very different relations between  $\text{NO}_3^-$  losses and particle ages in the different study areas (Figure 12c). Because first-order reaction kinetics was assumed, the fractional  $\text{NO}_3^-$  concentrations in Figure 12c are independent of specific  $\text{NO}_3^-$  input histories. At all of the sites except for Woodbury, denitrification had the potential to substantially reduce initial  $\text{NO}_3^-$  concentrations if there was sufficient ground water residence time in the denitrifying zones. Well construction or management practices that reduce water residence time in those zones, such as screening across confining layers, do not take full advantage of the natural  $\text{NO}_3^-$  attenuation capacity of the aquifer system. Sites like Woodbury do not have substantial  $\text{NO}_3^-$  attenuation capacity, but the short ground water residence times indicate that those types of systems would be relatively responsive to land use changes designed to reduce  $\text{NO}_3^-$  fluxes to the water table. These results illustrate that PSW vulnerability to  $\text{NO}_3^-$  contamination depends on complex variations and interactions between contaminant sources, reaction rates, transit times, mixing and perturbation of ground water flow in contrasting hydrogeologic settings.

## 5. Conclusions

[47] Understanding factors controlling the source and transport of  $\text{NO}_3^-$  to public supply wells (PSW) is challenging because of complex areas contributing recharge to the wells, spatial and temporal variability in  $\text{NO}_3^-$  sources within the contributing areas, spatially variable denitrification rates along flow paths leading to the wells, mixing, and possible perturbations in the flow system caused by well construction and (or) operation. Systematic studies of  $\text{NO}_3^-$  movement to PSW in four diverse hydrogeologic environments showed that  $\text{NO}_3^-$  in PSW was derived from varying proportions of agricultural (fertilizer, manure spreading), urban (septic leachate, fertilizer), and natural sources. Nitrate fluxes to the water table were larger in agricultural settings than in urban settings, indicating the PSW capture zones should be designed to limit inputs from agricultural sources. Denitrification in the aquifers was characterized by either slow rates in broad reaction zones within sandy fluvial and alluvial fan deposits or by fast rates in thin reaction zones at clay/sand contacts of glacial and marine deposits. In undisturbed flow systems, denitrification in the thin reaction zones provided more protection against  $\text{NO}_3^-$  contamination than the broad reaction zones. However, well construction and (or) operational features caused high- $\text{NO}_3^-$  water in two study areas to partially bypass or move more quickly through the denitrifying zones. In one sand aquifer, shallow  $\text{NO}_3^-$ -contaminated water bypassed the thin reaction zone and entered the producing interval of the PSW by moving down in local high-permeability features such as long well screens of nearby irrigation wells. In one carbonate aquifer, shallow  $\text{NO}_3^-$ -contaminated water bypassed the thin reaction zone and entered the producing interval of the PSW by moving through a high-permeability conduit. Consideration should be given to constructing and manag-

ing PSW to minimize bypass of denitrifying zones to take full advantage of the natural  $\text{NO}_3^-$  attenuation capacity of aquifer sediments.

[48] **Acknowledgments.** This work was funded by the USGS National Water-Quality Assessment (NAWQA) Program and National Research Program. Assistance in the stable isotope laboratory was provided by Tyler Coplen, Janet Hannon, Stan Mroczkowski, and Haiping Qi. We thank Janet Herman, Andrew Manning, James Tesoriero, Tom Torgersen, and two anonymous reviewers for comments on earlier drafts of this manuscript.

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## **Full Comments Section**

In general this report is well written and well organized, and provides a large amount of data that will be very useful to future studies and/or regulatory decisions. Most of the figures are easy to understand and well-integrated into the text. Appendix B presents a very good discussion of the uncertainties associated with the isotope data set for this study.

I do not see any major problems with the report, the majority of my comments are aimed at places in the report where additional clarification is needed and where I think uncertainties associated with some of the conclusions need to be either clarified or stated more strongly.

## **Answers to the review questions:**

- 1) In general, the purpose, scope and objectives of the project are clear. I'd suggest adding a couple of sentences in the relevant sections to indicate that the results of Phase I and Phase II are presented in more detail in other reports.
- 2) Reasons for selection of the various measurements are very clearly presented in the report.
- 3) The experimental design is clear, although I think there should be a bit more mention of the uncertainties associated with defining upgradient and downgradient wells (see specific comments below).
- 4) See general and specific comments about the isotope interpretations below. In general I think this is well presented, but a few edits & clarifications are still needed.
- 5) The conclusions are well supported by the results.
- 6) I don't see anything within the data that could more strongly link the nitrate contamination to specific sources. Based on the age dating results, isotope results, and fairly widespread occurrence of atrazine and a few other compounds, the nitrate in many of the wells is most likely from a mix of sources which would be challenging to tease apart, probably requiring a much more extensive sampling campaign and more knowledge of well depth and screen lengths.
- 7) In general the uncertainties are well addressed, I have added specific comments in places where I think some of the uncertainties either need to be more clearly stated, or need to be stated in the body of the report as well as at the end and/or in the appendices.

## **Isotope Data, General**

I think that some edits still need to be made regarding how the isotope data is presented in the body of the report. While Appendix B provides a very good discussion of the uncertainties associated with using isotopes to determine nitrate sources, within the body of the report there are several places where conclusive statements are made about nitrate sources to certain wells without the necessary explanations/caveats regarding the associated uncertainties. Within the discussions of many of the other compounds there

was better emphasis on what statements were the researchers' interpretations of the data, while there were several places where isotope interpretations were presented in ways that indicate definitive conclusions which I think over-reach the strength of the current data set. For example, on page 40 one sentence reads "For WW-04, the possible sources are fertilizer and animal waste, while the dominant source for WW-05 is animal waste". I think that this conclusion needs to be phrased differently to acknowledge the existing uncertainty- possibly use "WW-05 appears to be animal waste based on the elevated  $\delta^{15}\text{N}\text{-NO}_3$  values". Later on the same page the sentence "The isotopic data provide good evidence that animal waste is a dominant contributor to the nitrate contamination for WW-05" is a more accurate statement.

My primary concern regarding the isotope data is that due to limitations inherent in the study (well location, unknown depths, and unknown mixing/ flow paths, and potential mixed land use), the local end-member isotope values aren't well-defined, and potential effects of partial denitrification (which will increase isotope values while decreasing nitrate concentrations) couldn't be assessed for this study. I think the isotope data provide good indications of different sources of nitrate to different wells, and this is very well explained in Appendix B, but having what appear to be definitive statements about nitrate sources within the main body of the text could be very easily challenged by readers of the report.

**Some specific suggestions related to how the isotope data are reported:**

Within the body of the isotope data tables, the phrase "Fertilizer & Animal Waste" is used, even though in the table footnotes it is clarified that the interpretation is indeterminate- it could be all fertilizer, all animal, or a mixture. I suggest changing what is written in the table to "fertilizer and/or animal waste" which would be more accurate. The wells in which the higher  $\delta^{18}\text{O}\text{-NO}_3$  values suggest an atmospheric source are a bit more complicated- I think the most accurate phrasing might be "fertilizer and/or animal waste, with atmospheric contribution", since the  $\delta^{15}\text{N}\text{-NO}_3$  values are inconclusive, but the  $\delta^{18}\text{O}\text{-NO}_3$  values indicate that there has to be some sort of atmospheric contribution, either from specific fertilizers containing atmospheric nitrate, or from caliche leaching of long-term atmospheric nitrate deposition.

The overall isotope assessment of WW-02 should be changed to indeterminate- the nitrate concentration is slightly elevated in comparison to the lowest concentration wells, and there's no way to determine from the isotope & concentration data whether the nitrate came from soil cycling, or if there is simply a very small amount of either fertilizer or animal/human waste derived nitrate in the well. This does seem to be the only well where the isotopic composition and the concentration indicate that soil cycling can't be ruled out, but it isn't the only possible explanation.

**Other general comments:**

I think it would be very useful to include some brief comparisons between the different land uses for the distributions of the various compounds, which would be particularly valuable to future studies that might be interpreting well data in areas with very mixed land use. For example, are there any apparent trends in distributions of pesticides



between the different land uses? Did anything stand out as more frequently detected (or detected at higher concentrations) in the downgradient dairy wells in comparison to the downgradient septic and agricultural wells? It seems like atrazine was found across several different land uses- maybe include a comment about this and an brief evaluation of whether this means it might not be particularly helpful in future studies.

Approximately how long have the dairies been in operation? The age dating results suggest that many of the wells are probably sampling mixed older and younger water, so I'm still unsure about whether the upgradient vs. downgradient wells can be used to directly identify dairy-related impacts. I think this is addressed well in Section X, but I think the uncertainties associated with mixed land use need to be mentioned throughout the report.

If you anticipate controversy related to the release of this report, I think it might be worth mentioning early in the report that the dairies included in this study are expected to be fairly typical of dairy operations within the Yakima Valley, and that it is unlikely that the findings of elevated downgradient nitrate are unique to these particular dairies. However, I noticed that you said you selected dairies with over-application of fertilizers, so maybe they aren't typical. It would be worth some sort of very brief statement regarding whether you might expect conditions at these particular dairies to be applicable to other dairies in the area- I don't know how many dairies get flagged for over-application of N.

In the aerial photos of the Haak Dairy and the Dairy Cluster, it appears that there are a lot of agricultural fields surrounding the dairies. It also appears from the land use maps that there is some sort of agricultural land use (orchards? I can't quite tell if that's the same shade of pink in the land use map) upgradient of the dairies. I think there needs to be some discussion within the body of the report that the wells downgradient of the dairies may be drawing water from mixed agricultural land uses, or there needs to be a clearer presentation of why it is doubtful that surrounding fields that aren't dairy-related wouldn't be likely to contribute nitrate. Are the fields in the photos all/mostly related to the local dairies and received manure applications, how likely/ unlikely is it that nearby fields are getting synthetic fertilizers for non-dairy uses?

**Specific comments (in order that they appear in the report):**

Pg xi: UJ should be listed under acronyms and abbreviations, also "JN" is listed, but I didn't see it used anywhere in the table.

Pg 4. Last sentences of the introduction paragraph are somewhat confusing, since nitrate concentrations below 0.5 mg/L can often be found in non-impacted groundwater, and it is unclear if the 1.1 mg/L upper range might be applicable to this study area. Maybe rephrase to clarify.

Pg 4.Purpose & Scope- mention the possibility of synthetic fertilizers as nitrogen sources on dairies, since this is introduced as a unknown quantity later on in the report (unless there is additional information which makes it clear that fertilizers on the dairies must be

a small source of nitrate in comparison to the manure generated). Also, the report later on mentions that manure might be applied to non-dairy croplands.

Pg 7. Nitrogen Cycle- Somewhere in the last three paragraphs on the nitrogen cycle would be a good place to first mention that nitrate can build up in the soils of arid basins under certain circumstances.

Pg 9 Study Area- what is the possibility that some/ any of the wells sampled in this study are within the deeper aquifer? This section goes into detail about the uncertainties, but in the sections on the different well results, the report makes it sound like there is reasonable certainty that the upgradient and downgradient wells are tapped into an aquifer with a known and somewhat rapid flow direction. Since approximate depth to the confined aquifer isn't mentioned in the Study Area section, I can't evaluate the likelihood that any of the wells might be in a deeper aquifer with an unknown flow direction.

Pg 11. It would be helpful to explicitly state that the details of Phase I and Phase II are/will be included in separate reports. When I first read this section it was difficult to tell if this was supposed to be a comprehensive reporting of all Phases, or just Phase III, with summaries of the other two phases.

Pg11. Since Figure 6 doesn't have a caption, the detailed explanation (which now seems to be the second to last paragraph in the Irrigated Cropland subsection) should be moved close to the first mention of Figure 6. At first I wasn't able to figure out how the estimates were derived or what the figure really represented because the explanation was much further along in the section

Pg 11-12 Section goes from Figure 6 to Figure 8, Figure 7 doesn't appear to be mentioned until pg 14- change the order of the figures?

Pg 13. Where does the water discharge from these WWTPs get released? Does it leave the area in surface water, or is it used along with some of the biosolids in land application. If the WWTPs only go to secondary treatment, this could be a source of high N water somewhere in the study area. Briefly mention why this either might be important or explain why it can be ignored for this study (discharge to surface water, tertiary treatment with low final N levels, etc).

Pg 14. Is the Phase II data being evaluated in a separate report? It seems like this dataset would be useful for an analysis of land use vs. nitrate concentrations, but it is difficult to tell from the way this section is written whether or not additional evaluations of the Phase II dataset were done somewhere else.

Pg 15. How certain is it that the dairy areas only contain dairy-related sources? The aerial photos seem to show surrounding fields- are these all dairy fields? Maybe state that within the mixed septic & irrigated cropland areas, these sources were all within X kilometers of the sampled wells, while in the dairy areas, only dairy-related fields were located within X kilometers of the sampled wells.

Pg 16. How far away were other potentially significant land uses from the wells with potential septic influence? Are they reasonably isolated from fertilizer & irrigated fields?

Pg 16. The final paragraph of the Phase 3 section is a bit confusion, particularly because of the sentence that starts “The five groups include:...” but then only lists three categories. Rephrase for clarity.

Pg 17: In the criteria for selection of septic systems, there needs to be further explanation about how well the selected wells met the criteria of “minimal upgradient nitrate sources other than septic.” Maybe an approximate distance away from any irrigated and fertilized fields? On page 15, it is mentioned that the three other areas contain both septic systems and irrigated cropland sources.

Pg 21. Microbiology section- it would be easier to follow if the report indicated which samples were tested using MST, rather than listing which samples were not tested.

Pg 24. The discussion of the veterinary pharmaceuticals is somewhat confusing. One sentence states that “Detections of the compounds in Table 3 in water wells would provide evidence that dairies are a likely source of those compounds”, but then further reading makes it sound as if this is not necessarily true since some of the compounds are also used in humans, and the report cites two references saying that some of these compounds have been detected in wastewater treatment facilities. Is it only some of these compounds that can rule out human-derived waste? When other researchers detected these compounds in the wastewater treatment plants, did they conclude that somehow animal waste was entering the treatment facilities?

Pg 26. Second paragraph in Isotopic Analysis. I would suggest changing the phrasing of this paragraph to state that multiple studies have shown that various dominant nitrate sources can have overlapping isotopic compositions, and that unless the range of isotopic values for nitrate source end-members within the local study area has been well measured (and have distinct isotope values), nitrate which falls into the isotopic range in which the sources overlap cannot be distinguished based on isotopes alone. The phrase “the science is still evolving” is somewhat true, but it implies that additional understanding of the fractionations will eventually allow us to distinguish these sources. This isn’t really true- the area of overlap/ undetermined isotope composition has been found in multiple studies, and it is extensive local sampling (including well-constrained end-members) within a single study area, not additional general scientific understanding, that has the potential to allow different sources to be separated with more confidence. I think it is important to make brief mention of how in-situ denitrification and/or mixing can shift the isotope values away from the original source signal. This is fairly well addressed in Appendix B, but I think it needs to be specifically mentioned within the body of the report. Also, some of the wells did have low field values for dissolved oxygen (I’m not too sure how accurate the field measurements are considered to be for this study), so shifts in individual wells due to partial denitrification is a possibility. However, I looked

at the plots of dissolved oxygen vs  $\delta^{15}\text{N-NO}_3$  and did not see any trend, so I think it is reasonable to rule out in-situ denitrification as a significant factor.

Pg 27. Specify that the atmospheric  $\delta^{18}\text{O}$  of 23.5 per mil is for  $\text{O}_2$  gas (to differentiate it from  $\text{NO}_x$  in the atmosphere).

Pg 28. Not too sure what is meant by “groundwater elevations fluctuate rapidly” - is this fluctuations in depth to groundwater, or rapid travel along a flow path from high to low elevations?

Pg 28. I think it would be a good idea to include a statement about whether or not, or to what extent, the reported age dates can be considered valid. Does the presence of over value samples suggest that all the samples in the area could be altered by local geology, or is this something that would only impact the wells that are already flagged as having invalid  $\text{SF}_6$  ages?

Pg 29. What is the function of the Dairy A ditch? Since it is mentioned, indicate whether it carries waste, or just surface water for transport, irrigation, some other use? Is it lined or unlined- I’m wondering if it could leak in a similar fashion to the lagoons and also serve as a local source of rapidly infiltrating water.

Pg 29. The pivot-irrigated fields are clearly visible in the aerial photos- do the other fields around the pivot also receive liquid or solid manure?

Table 5: There seems to be at least a couple of things wrong with the reported values for the lagoons- it should be L-01, -02, and -03 (the table lists L-03, -04, and -05), but the Ammonia values in the table appear to be from L-04, -05, and -06 according to Table A2 in the Appendix.

Pg 32. Paragraph directly following Table 6- Is there a reference for the barium and zinc levels reported for local surface waters?

Pg 33-34. Since atrazine was found in the upgradient well and the downgradient wells, it seems like you should mention that the atrazine could also be from other surrounding land uses, the occurrence of atrazine in the application fields doesn’t provide a very strong link to the downgradient wells, particularly since it seems to have been found in wells with different land uses as well.

Pg 41. It’s unclear what the differences are between the dairy supply wells and the domestic drinking water wells. Is there evidence that well construction/ depth/ pumping rates are different, or is the only known difference the choice of use (drinking vs. other uses)? Could the supply wells be considered upgradient or mid-gradient?

Pg 44. The lagoons are mentioned as a potential source for the major ions, but is there any evidence that the lagoons are more important than the sprayfields? If they are using the spray fields partially for liquid manure disposal (instead of balanced entirely for

limited fertilization), it seems like the large areas covered by the sprayfields could be equally as important as the lagoons.

Pg 44. “However, elevated levels of perchlorate were seen in only two wells”- include which wells were elevated, and what the threshold was for defining elevated.

Pg 47. Since the tetracycline was detected in higher levels in the upgradient wells in comparison to the downgradient wells, it seems like it could be coming from mixing along flowpaths with other sources, and not necessarily from the dairies. I think it is quite reasonable to state that the dairy cluster is a possible source, but it seems like the other possibilities should also be clearly stated here.

Pg 48. Dairy cluster- hormones. The upgradient well (WW-06) isn’t mentioned as either having hormone detections or not having hormone detections.

Pg 48. first sentence of Dairy cluster- isotopic analysis, chose a different word instead of “completed”, maybe use “performed” or “attempted”, since in the first sentence it says that isotope analysis was completed on all wells, then the next sentence says it couldn’t be completed on certain wells.

Table 15. In the table, change to fertilizer and/or animal waste for consistency and accuracy.

Pg 49. Paragraph following Table 15- change “dominant source is animal waste” to something like dominant source appears to be animal waste, or “ $\delta^{15}\text{N}$ -NO<sub>3</sub> values indicate that the dominant nitrate source is most likely animal waste”.

Pg 50. The sentence “Table 16 indicates the source of nitrate in the dairy lagoons is animal waste” seems very unclear. Maybe change this to something like “Table 16 provides an assessment of the level of volatilization of the animal waste as indicated by the isotope values”.

Pg 54. 2<sup>nd</sup> paragraph- need qualifiers on the isotope assessments, the isotope values indicate or suggest the different sources for the different wells, replace “is” with some phrasing that incorporates the uncertainty, or make it clear that the assigned sources are and interpretation based on the evidence.

Pg 54. 3<sup>rd</sup> paragraph- this says “upgradient wells” but it appears there is only one upgradient well?

Pg 54. 4<sup>th</sup> paragraph- the phrase “ dominant nitrate source for the pharmaceuticals” doesn’t make sense, I’m not too sure what the paragraph is trying to convey.

Pg 54. It’s unclear what the sample SP-04 represents, maybe remove it completely from the Appendix tables if it isn’t part of the report?

Pg 55. Major ions- I don't think the use of major ions has to be limited to transformations along a flowpath. As you mention earlier in the report (in regards to ion ratios), major ion distributions can also sometimes be indicative of specific water and/or contaminant sources.

Pg 55. Septic Systems- trace elements. It would be interesting to add a few sentences comparing the distributions of trace elements found in these wells with the distributions found in the downgradient dairy wells, even if the conclusion is that they aren't helpful at this time for distinguishing different contaminant sources.

Pg 61. 2<sup>nd</sup> paragraph. I suggest using animal/human waste instead of just "animal waste". It is stated earlier in the report that animal waste includes humans, but I think clarifying this again will help avoid confusion for people reading the report.

Pg 62. It would be interesting to include a paragraph explicitly comparing the various N concentrations in the field soils with those found in the dairy soils.

Pg 63. Change "only two were detected in the six water wells" to "only two were detected in ANY of the six water wells" in order to clarify that there weren't detections in all six wells.

Ph 63. Pharmaceutical, 1<sup>st</sup> sentence- change "Nine compounds were detected" to "nine compounds were detected in this well".

Pg 64. Hormone- right after the bulleted list. This sentence is a bit unclear, it says that the hormones are naturally produced by animals, and can be expected in septic systems- does this mean that they are also produced by humans, or do non-human animal produced hormones often reach septic systems. Even though the term "animals" technically includes humans, I'd suggest specifying throughout the report which things are non-human animals only, and which things are human & other animals.

Pg 65. 1<sup>st</sup> paragraph. I'd suggest including one sentence clarifying that it is the elevated d18O-NO<sub>3</sub> which indicates an atmospheric contribution.

Pg 66. Were these the only wells where bentazon was detected?

Pg 66. What is the approximate distance of the dairy?

Pg 67. Would it be possible to include a general land use assessment for wells 18 and 30. Are they in predominantly residential areas, irrigated croplands, something else?

Pg 67. I think the complications associated with mixed land uses should also be mentioned here, particularly because the detections of some compounds in the various upgradient wells indicates the presence of different sources of various compounds within the study areas. You can emphasize that sites were selected to try and minimize mixed

land use, but it will always be close to impossible to find areas with only a single land use.

Pg 69. Give a very brief summary of why the microbial and age dating data weren't useful in identifying sources.

Pg 69. The sentence "the presence of these compounds in theses sources..." is a bit unclear, maybe change to "the presence of these compounds in the primary sources (lagoons, manure piles, soils) but not in the water wells..."

Appendix Table A1: SP-04 seems to be missing from this table. It appears in some tables but not in others?

Appendix Table A3: what does the qualifier KK mean for sample SO-01 NO<sub>3</sub>N/Total solid? KK isn't listed in the Acronyms/ Abbreviations section.

## Questions

1. Are the purpose, scope, and objectives of the project clear?

**Response:** The purpose of the study is clearly stated at the beginning of the document (1st paragraph in the Introduction). The placement of the purpose at this location is vital to making the document clear to the reader. The scope of the study is also presented clearly in the first paragraph.

2. Is it clear why we selected certain chemical classes (e.g., hormones) or analytical techniques (e.g., isotopic analysis) to serve as potential tracers for nitrate contamination?

**Response:** Overall your explanation of chemical classes is clear. The list of hormones in Table 4 indicates which animals the hormones are used for. There is no mention of any being used in dairy cows. Is this correct? The first 5 listed under “Analyzed at UNL Only” do not indicate for which animals they are from. I am assuming they are associated with mammals in general.

It would be valuable to have some information on the mobility and decay rates of the hormones and pesticides in soils. If an organic constituent is not mobile it is unlikely that they will move to ground water, thus not serve as good indicators of nitrate sources. Need to make sure the selected organic constituents have a purpose not just “this is what the lab can analyze for so we will include” justification behind selection of constituents.

3. Is the experimental design clear?

**Response:** In the main text the design is clear but a few more specifics could be included. These suggestions are listed in other responses.

Regarding Appendix C, some terms need to be better defined – qualified and out of control. When I read “qualified estimated” I think a “scientific guess”. Guessing has no place in scientific investigations. I am assuming that this is not the case, so a better explanation of what “qualified estimates” are would be good. References to QA procedures from other reports are plentiful in this Appendix. Many readers will not search for these so it may be good to summarize them in this report.

After reading Appendix C I had the impression there were a lot of issues with organic compound sample analysis that made me question the validity and need of the data.

4. Is the approach taken for evaluating the isotopic data reasonable given the results from the study and the literature on isotopic analysis (e.g.,  $\delta^{15}\text{N}$ - $\text{NO}_3$  water well values greater than 8.4‰ characterized as dominated by animal waste;  $\delta^{15}\text{N}$ - $\text{NO}_3$  water well values less than 2.0‰



characterized as dominated by fertilizer; and  $\delta^{15}\text{N}$ - $\text{NO}_3$  water wells values between 2.0‰ and 8.4‰ being characterized as isotopically in determinant as to animal waste and/or fertilizer).

**Response:** I am not an expert on isotope analysis but based on what I do know, your approach seems reasonable. Listing the shortcoming of the analysis and what conclusions can be drawn was a good strategy. I would recommend placing the information found in the footnotes of Table 8 into the design section for “D. Isotopic Analysis” on page 27.

5. Are the conclusions supported by the results?

**Response:** The conclusions are supported by the data.

6. Are there results which could be more strongly used to link nitrate contamination to sources?

7. **Response:** Not that I could see.

8. Are the uncertainties adequately addressed and clearly articulated?

9. **Response:** Yes.

#### **Other Comments:**

When giving nitrate concentrations make sure to specify wheather the concentrartios are reported as  $\text{NO}_3$  or  $\text{NO}_3\text{-N}$  (e.g. Background section, Page 5: “...0.5 mg/L and to 1.1 mg/L...”)

In Figure 1, some leaching path arrows look line runoff paths.

Including Section X. Study Limitations and Uncertainties was important. Regarding the second paragraph discussion on samples from wells with high nitrate and low oxygen, denitrification will not proceed under these situation unless there is an energy source (usually organic C) for the denitrifying bacteria to use and a terminal electron acceptor ( $\text{NO}_3^-$  in this case due to lack of a better electron acceptor,  $\text{O}_2$ ). These waters could have been low in organic C thus accounting for the high levels of nitrate with low oxygen in the water. Did you measure organic C content of the water? This point (requirement of an energy source) was missing in the introduction section dealing with denitrification.

**Submit Time:** 8/31/2011 18:13:23  
**From:** CN=Stephen Kraemer/OU=ATH/O=USEPA/C=US  
**To:** CN=JohnM Johnston/OU=ATH/O=USEPA/C=US@EPA  
**Cc:** CN=Candida West/OU=ATH/O=USEPA/C=US@EPA  
**Subject:** Fw: Yakima Groundwater: Third Party Reviewers

I believe this is the call that you were talking about. Roseanne left a message for me, and then I left a message for her. Good she talked to Candida. I bet a team of ERD folks could provide the review. The other person might be Mohamed Hantush of NRMRL.

----- Forwarded by Stephen Kraemer/ATH/USEPA/US on 08/31/2011 02:11 PM -----

From: Roseanne Lorenzana/R10/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Roy Sidle/ATH/USEPA/US@EPA, Candida West/ATH/USEPA/US@EPA, Stephen Kraemer/ATH/USEPA/US@EPA  
Date: 08/30/2011 04:55 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

Mike:

In case you didn't get my voicemail, the ORD National Exposure Research Lab contacts:  
Roy Sidle, Division Director 706-355-8001  
Candida West 706-355-8023  
Steve Kraemer 706-355-8340

These folks will help pin down exactly who can help with the review.  
Roseanne

-----  
Roseanne M. Lorenzana, DVM, PhD  
Diplomate of the American Board of Toxicology  
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<http://intranet.epa.gov/ospintra/scienceportal/htm/regionalscience.htm>

Michael Cox---08/23/2011 02:30:31 PM---Yeah I would say someone should be able to review in two-days. Michael Cox Office of Environmental A

From: Michael Cox/R10/USEPA/US  
To: Roseanne Lorenzana/R10/USEPA/US@EPA  
Cc: Curt Black/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
Date: 08/23/2011 02:30 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

Yeah I would say someone should be able to review in two-days.

Michael Cox

Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Roseanne Lorenzana---08/23/2011 02:26:33 PM---Hi Mike - I have been checking around. Discussion typically comes to the question of how many work

From: Roseanne Lorenzana/R10/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Curt Black/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
Date: 08/23/2011 02:26 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

Hi Mike - I have been checking around. Discussion typically comes to the question of how many work hours are involved. Are you able to estimate? Would 16-24 hours be a reasonable estimate?

Roseanne

---

*Roseanne M. Lorenzana, DVM, PhD, DABT  
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<http://intranet.epa.gov/ospintra/scienceportal/htm/regionalscience.htm>*

Michael Cox---08/23/2011 02:16:29 PM---Roseanne,  
**Ex. 6 Personal Privacy (PP)**

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From: Michael Cox/R10/USEPA/US  
To: Roseanne Lorenzana/R10/USEPA/US@EPA  
Cc: Sheila Fleming/R10/USEPA/US@EPA, Curt Black/R10/USEPA/US@EPA  
Date: 08/23/2011 02:16 PM  
Subject: Yakima Groundwater: Third Party Reviewers

Roseanne,

I was checking if you had a chance to see if anyone from ORD maybe interested in participating in a third party review for the Yakima Report?

Thanks.

Michael Cox  
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US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov



**Subject:** Re: Yakima Groundwater: Third Party Reviewers  
**To:** CN=Michael Cox/OU=R10/O=USEPA/C=US@EPA  
**Cc:** CN=Roy Sidle/OU=ATH/O=USEPA/C=US@EPA CN=Candida West/OU=ATH/O=USEPA/C=US@EPA CN=Stephen Kraemer/OU=ATH/O=USEPA/C=US@EPA  
**From:** CN=Roseanne Lorenzana/OU=R10/O=USEPA/C=US  
**Submit Time:** 8/30/2011 20:55:08

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<http://www.epa.gov/region10/>  
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Cc: Curt Black/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
Date: 08/23/2011 02:30 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

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Date: 08/23/2011 02:26 PM  
Subject: Re: Yakima Groundwater: Third Party Reviewers

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<http://intranet.epa.gov/ospintra/scienceportal/htm/regionalscience.htm>*

Michael Cox--08/23/2011 02:16:29 PM--Roseanne

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From: Michael Cox/R10/USEPA/US  
To: Roseanne Lorenzana/R10/USEPA/US@EPA  
Cc: Sheila Fleming/R10/USEPA/US@EPA, Curt Black/R10/USEPA/US@EPA  
Date: 08/23/2011 02:16 PM  
Subject: Yakima Groundwater: Third Party Reviewers

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**Subject:** Yakima Report: Schedule  
**To:** CN=Thomas Eaton/OU=R10/O=USEPA/C=US@EPA CN=Marie Jennings/OU=R10/O=USEPA/C=US@EPA CN=Sheila Fleming/OU=R10/O=USEPA/C=US@EPA CN=Eric Winiecki/OU=R10/O=USEPA/C=US@EPA CN=Chan Pongkhamsing/OU=R10/O=USEPA/C=US@EPA CN=Jennifer MacDonald/OU=R10/O=USEPA/C=US@EPA CN=Caryn Sengupta/OU=R10/O=USEPA/C=US@EPA CN=Rochelle Labiosa/OU=R10/O=USEPA/C=US@EPA CN=Curt Black/OU=R10/O=USEPA/C=US@EPA Cox.Michael@epa.gov CN=Sandra Halstead/OU=R10/O=USEPA/C=US@EPA CN=Gina Grepo-Grove/OU=R10/O=USEPA/C=US@EPA CN=James Lopez-Baird/OU=R10/O=USEPA/C=US@EPA  
**Cc:** CN=Jeff Philip/OU=R10/O=USEPA/C=US@EPA CN=Wenona Wilson/OU=R10/O=USEPA/C=US@EPA  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 7/25/2011 17:56:48

Okay. Here is my attempt at a realistic timeframe for the Yakima Report. Probably not as quick as anyone would want.

August 10th: Draft for internal EPA review

August 24th: Comments due from internal review

September 12th: Draft to third party review

October 3rd: Third party review comments due

October 17th; Draft Final for internal EPA review

October 24th: Final comments due from internal EPA review

November 14th: Final report

We can discuss at the next meeting or via email.

Michael Cox  
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cox.michael@epa.gov

**Subject:** Yakima Report: Schedule - Update  
**To:** CN=Michael Cox/OU=R10/O=USEPA/C=US@EPA  
**Cc:** CN=Caryn Sengupta/OU=R10/O=USEPA/C=US@EPA CN=Chan Pongkhamsing/OU=R10/O=USEPA/C=US@EPA CN=Curt Black/OU=R10/O=USEPA/C=US@EPA CN=Eric Winiacki/OU=R10/O=USEPA/C=US@EPA CN=Gina Grepo-Grove/OU=R10/O=USEPA/C=US@EPA CN=James Lopez-Baird/OU=R10/O=USEPA/C=US@EPA CN=Jeff Philip/OU=R10/O=USEPA/C=US@EPA CN=Jennifer MacDonald/OU=R10/O=USEPA/C=US@EPA CN=Marie Jennings/OU=R10/O=USEPA/C=US@EPA CN=Rochelle Labiosa/OU=R10/O=USEPA/C=US@EPA CN=Sandra Halstead/OU=R10/O=USEPA/C=US@EPA CN=Sheila Fleming/OU=R10/O=USEPA/C=US@EPA CN=Thomas Eaton/OU=R10/O=USEPA/C=US@EPA CN=Wenona Wilson/OU=R10/O=USEPA/C=US@EPA  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 8/9/2011 23:00:19

Just an update on the schedule for the report. For the following reasons I am proposing we set the end of the calendar year 2011 as the date for the final report.

1. Based on feedback from the team, we will need more time for internal review, comments, and discussion on the report than I originally scheduled. This is compounded by summer vacation schedules.
2. We continue to work with UNL to try and bring to a close the evaluation of the data from their lab. We are close, but still have a few questions for UNL that we need addressed.
3. We are working with USGS on the interpretation of the isotopic data. This has been very useful, but the person we are working has other work she needs to do. I think the isotopic data is critical and we need to make sure we are interpreting correctly.

### **Proposed Schedule**

August 23rd: Draft for internal EPA review

September 23th: Comments due from internal EPA review (assumes we will have a couple of meetings to discuss report)

October 7th: Draft to third party review

October 28th: Comments from third party review

November 18th: Draft final report for review

December 2nd: Final comments

December 31st: Final Report

Any comments on schedule please call, email, or stop by.

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206-553-1597  
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Michael Cox---07/25/2011 10:56:49 AM---Okay. Here is my attempt at a realistic timeframe for the



Yakima Report. Probably not as quick as

From: Michael Cox/R10/USEPA/US  
To: Thomas Eaton/R10/USEPA/US@EPA, Marie Jennings/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA, Eric Winiecki/R10/USEPA/US@EPA, Chan Pongkhamsing/R10/USEPA/US@EPA, Jennifer MacDonald/R10/USEPA/US@EPA, Caryn Sengupta/R10/USEPA/US@EPA, Rochelle Labiosa/R10/USEPA/US@EPA, Curt Black/R10/USEPA/US@EPA, Cox.Michael@epa.gov, Sandra Halstead/R10/USEPA/US@EPA, Gina Grepo-Grove/R10/USEPA/US@EPA, James Lopez-Baird/R10/USEPA/US@EPA  
Cc: Jeff Philip/R10/USEPA/US@EPA, Wenona Wilson/R10/USEPA/US@EPA  
Date: 07/25/2011 10:56 AM  
Subject: Yakima Report: Schedule

---

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Michael Cox  
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**Subject:** Yakima: Science Inventory  
**To:** CN=Sheila Fleming/OU=R10/O=USEPA/C=US@EPA CN=Jennifer MacDonald/OU=R10/O=USEPA/C=US@EPA  
**Cc:** Cox.Michael@epa.gov  
**From:** CN=Michael Cox/OU=R10/O=USEPA/C=US  
**Submit Time:** 12/18/2012 20:26:47

I was going to forward to Gina, but first wanted to make sure we all agree with suggestions.

---

Gina,

I reviewed the Science Inventory and have the following suggestions.

Description: Results of a study EPA conducted to investigate the contribution of various sources to the high nitrate levels in groundwater **and residential drinking water wells** in the Lower Yakima Valley of Washington.

Purpose/Objective: Data report

URLs/Downloads (suggest the following order and suggest not using caps)

Relation between Nitrates in .... (needs complete title - if it is long then I would suggest using Lower Yakima Valley Study)

Third Party Review Panel

Third Party Review Charge

Comments from Stephen Kraemer, PhD, USEPA and Roger Burke, PhD, USEPA

Comments from Lorraine Edmond, USEPA

Comments from Megan B. Young, PhD, USGS

Comments from David Tarkalson, PhD, USDA

Thanks.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

Gina Grepo-Grove---12/18/2012 08:03:12 AM---Holo Mike and Sheila, I filed an episodic flexiplace today.

**Ex. 6 Personal Privacy (PP)** I'm sup

From: Gina Grepo-Grove/R10/USEPA/US  
To: Michael Cox/R10/USEPA/US@EPA  
Cc: Sheila Fleming/R10/USEPA/US@EPA  
Date: 12/18/2012 08:03 AM  
Subject: Re: Yakima: FOIAs

Holo Mike and Sheila,

I filed an episodic flexiplace today. **Ex. 6 Personal Privacy (PP)** I'm supposed to be on regular flexiplace tomorrow but if the road will be clear I will be in also on Thursday.

Ginna Grepo-Grove

R10 Quality Assurance Manager  
(206) 553-1632 - Office Phone Number  
(206) 553-8210 - Fax Number

-----Michael Cox/R10/USEPA/US wrote: -----

To: Gina Grepo-Grove/R10/USEPA/US@EPA, Sheila Fleming/R10/USEPA/US@EPA  
From: Michael Cox/R10/USEPA/US  
Date: 12/17/2012 04:13PM  
Subject: Yakima: FOIAs

Gina,

Wanted to see what your plans were for this week. We need to get a FOIA out on the data and wanted to see if you plan on being around this week so we can meet to go over one more time.

Thanks.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

## Appointment

---

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 2/22/2013 11:12:13 PM  
**To:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]; Pirzadeh, Michelle [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]; Holsman, Marianne [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]; Stern, Allyn [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=2e2d6f5df08d4e9abbff2c0880407775-Stern, Allyn]; Kelly, Joyce [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=B1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Kowalski, Ed [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=05111ec2f4314434a376f1c84c851820-Kowalski, Ed]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]

**Subject:** Yakima Well Report - Comment Review

**Location:** 14th Floor - ET Conf Room; Meet-me-line

Ex. 6 Personal Privacy (PP)

**Start:** 2/25/2013 4:00:00 PM

**End:** 2/25/2013 4:30:00 PM

**Show Time As:** Busy

**Importance:** High

Dennis McLerran  
Michelle Pirzadeh  
Marianne Holsman  
Bill Dunbar  
Allyn Stern  
Joyce Kelly  
Ed Kowalski  
Tom Eaton

Monday, Feb 25, 8:00-9:00 am.

Topic is Yakima Well Report Review Comments

Message

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**From:** Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10FA13022096442D83A97A2B7A699F06-EATON, THOMAS]  
**Sent:** 3/4/2013 4:22:01 PM  
**To:** Cox, Michael [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=cddd6a5bb3c2477183799ef56cdb464f-Cox, Michael]; Winiecki, Eric [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=45fa974e305b49cd8af513001a800543-Winiecki, Eric]; MacDonald, Jennifer [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=507c2f57e60741b2b137cdf441c1c731-MacDonald Jennifer]; Fleming, Sheila [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=5d3e29dad0ec4cd98d87e1a142435c57-Fleming, Sheila]; Fuentes, Rene [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=3eb2a2e594934952b91ca7ade37262c5-Ruentes, Rene]; Jennings, Marie [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=ec4bd403e1994b80976542bb9458ae72-Jennings, Marie]  
**Subject:** RE: Updated Lagoon Section  
**Attachments:** Yakima Comments Lagoon March 1 2013.docx

Looks good - a few minor edits

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**From:** Cox, Michael  
**Sent:** Friday, March 01, 2013 4:54 PM  
**To:** Winiecki, Eric; Eaton, Thomas; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie  
**Cc:** Cox, Michael  
**Subject:** RE: Updated Lagoon Section

If you want to comment on the lagoon section please use this version. It is in the right format and will make easier to insert. I combined a few of the comments and responses but no change in the content.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
[cox.michael@epa.gov](mailto:cox.michael@epa.gov)

---

**From:** Winiecki, Eric  
**Sent:** Friday, March 01, 2013 4:10 PM  
**To:** Cox, Michael; Eaton, Thomas; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie  
**Subject:** Updated Lagoon Section

Hi folks,

I incorporated Tom's comments into the lagoon responses and made some additional improvements. Please review this version. The second file above is an attachment that is referenced in one of the responses.

Thanks,

Eric

Message

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**From:** Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10FA13022096442D83A97A2B7A699F06-EATON, THOMAS]  
**Sent:** 3/26/2013 9:10:08 PM  
**To:** esanche@yakama.com [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=dc004cb3b2e645a7a035781fcb60afcc-esanche@yakama.com]; Phil Rigdon [prigdon@yakama.com]  
**Subject:** Yakima documents

All comments we received on the report, responsiveness summary, errata sheet of changes to the report and a new report summarizing the data collected at the 10 wells we drilled in the vicinity of the dairies. I believe the new data report addresses several of the technical comments we received. Call if you have questions.

<http://yosemite.epa.gov/r10/water.nsf/GWPU/lyakimagw>

Thomas Eaton  
Director, Washington Operations Office  
360-753-8086  
206-295-9364 (cell)

## Appointment

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**From:** Magorrian, Matthew [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=A25BC04CD8B54A5E9A85488AC486413C-MAGORRIAN, MATTHEW]  
**Sent:** 2/22/2013 11:12:08 PM  
**To:** Pirzadeh, Michelle [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]; Holsman, Marianne [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]; Stern, Allyn [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=2e2d6f5df08d4e9abbff2c0880407775-Stern, Allyn]; Kelly, Joyce [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=B1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Kowalski, Ed [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=05111ec2f4314434a376f1c84c851820-Kowalski, Ed]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]  
**Subject:** Yakima Well Report - Comment Review  
**Location:** 14th Floor - ET Conf Room; Meet-me-line: Ex. 6 Personal Privacy (PP)  
**Start:** 2/25/2013 4:00:00 PM  
**End:** 2/25/2013 4:30:00 PM  
**Show Time As:** Busy  
**Importance:** High

Message

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**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 2/15/2013 12:40:15 AM  
**To:** Sussman, Bob [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=E85454566bd04951a8b08dcc9df57b4c-Sussman, Bob]; James O'Hara [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=F2982e84e36742b585369f33356e1625-James O'Hara]  
**CC:** Holsman, Marianne [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kowalski, Ed [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=05111ec2f4314434a376f1c84c851820-Kowalski, Ed]; Kelly, Joyce [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=B1487930f7634eecebe7c90bbd3003599-Kelly, Joyce]  
**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS  
**Attachments:** Yakima ltr to NRCS 2-14-13ver3.docx



Yakima ltr to  
NRCS 2-14-13ve...

Bob and Jim:

Here is a first cut at a letter from me to Washington NRCS Director Roylene Rides-at-the-Door.

Dennis



Message

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**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 3/21/2013 9:14:28 PM  
**To:** Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]  
**CC:** Magorrian, Matthew [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=a25bc04cd8b54a5e9a85488ac486413c-Magorrian, Matthew]  
**Subject:** Re: Yakima: Lagoon seepage

Dan:  
Let's discuss on the call.  
Dennis

---

**From:** Opalski, Dan  
**Sent:** Thursday, March 21, 2013 4:51:46 PM  
**To:** McLerran, Dennis  
**Cc:** Magorrian, Matthew  
**Subject:** FW: Yakima: Lagoon seepage

Dennis --

The attachment is the targeted revision to the lagoon seepage lanaguage in the report itself. I think Mike and Sheila have done a good job of matter-of-factly citing to NRCS (and other) sources for actual information, but being careful not to attribute to them any of our own calculations. Please take a look and see what you think. I'm not thinking this is something we would need to loop back through NRCS on, but I want to get your read because that will mean another step of potentially undefined duration.

By the way, you may notice (I did) that a couple of the citations are now to "NRCS Washington" as opposed to "NRCS." This apparently is the more accurate citation, and reflects that the individual state office develop state-specific practices, consistent with the notion that they incorporate what is in the particular State's own standards and requirements.

We can add this to our 3:30 agenda or take it off line, as appropriate. DanO.

---

**From:** Cox, Michael  
**Sent:** Thursday, March 21, 2013 11:22 AM  
**To:** Opalski, Dan  
**Cc:** Fleming, Sheila; MacDonald, Jennifer; Winiecki, Eric; Jennings, Marie; Cox, Michael  
**Subject:** Yakima: Lagoon seepage

Dan,

I have attached the revised sections that discuss the lagoon seepage and also left a copy on your chair.

We are in the Kenai if you have time to discuss.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597

cox.michael@epa.gov

Message

---

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 3/1/2013 4:22:19 PM  
**To:** Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]  
**Subject:** Re:

Thanks Michelle

---

**From:** Pirzadeh, Michelle  
**Sent:** Friday, March 01, 2013 11:04:44 AM  
**To:** McLerran, Dennis  
**Subject:** Re:

Dennis,

I've been in touch with Tom and Dan. We've gathered all of the documents and Tom is sending them out from WOO within the hour.

Michelle Pirzadeh  
Deputy Regional Administrator  
U.S. Environmental Protection Agency, Region 10  
1200 Sixth Avenue, Suite 900  
Seattle, Washington 98101  
(206) 553-1234

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:57:44 PM  
**To:** Pirzadeh, Michelle  
**Subject:** FW:

Michelle:

Glenn is asking us to Fed Ex him copies of the documents I identify below. He does not have printing capability so he needs hard copies. The address to Fed Ex them to is:

Glenn Paulson

Ex. 6 Personal Privacy (PP)

Ex. 6 Personal Privacy (PP)

If we need to reach him for an address for next week his cell number is Ex. 6 Personal Privacy (PP) Thanks!

Dennis

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:58 PM  
**To:** Paulson, Glenn  
**Cc:** Sussman, Bob; Pirzadeh, Michelle  
**Subject:**

Glenn:

Thanks for talking with me this afternoon. I am confirming that we agreed that a release of the additional well data to the attorney for the citizens' group next Wednesday is OK with you. I will also Fed Ex you a copy of the narrative drafted for the additional data, a copy of the original narrative, a copy of the NRCS comment letter and a copy of the Dairy Federation comment letter.

Since we spoke, I have learned that the attorney for the citizens' group has not agreed to hold off on filing a complaint regarding his FOIA request on the additional well drilling data. So, we will be responding to the press if he goes public with a press release. We have triggered a 10 day extension under the FOIA regulations as we complete our review of the additional data. I still believe it will be appropriate to just release the data to him next Wednesday and follow later with the narrative if needed.

*Dennis J. McLerran  
Regional Administrator  
U.S. EPA, Region 10*

*Office: 206-553-1234  
Fax: 206-553-1809*

Message

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 3/1/2013 5:32:45 PM  
**To:** Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]  
**CC:** Hockenbury, Laura [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=ccf5c33e87eb46168e43d4fe8b5bb9ba-Hockenbury, Laura A.]; Kelly, Joyce [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=b1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Stern, Allyn [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2e2d6f5df08d4e9abbff2c0880407775-Stern, Allyn]  
**Subject:** Re: Fw:

Yes. I just don't know if je will be done or have suggested revisions. I've asked him to review it

Ex. 6 Personal Privacy (PP)

Ex. 6 Personal Privacy (PP)

Dennis

---

**From:** Opalski, Dan  
**Sent:** Friday, March 01, 2013 12:30:42 PM  
**To:** McLerran, Dennis; Eaton, Thomas; Pirzadeh, Michelle  
**Cc:** Hockenbury, Laura; Kelly, Joyce; Stern, Allyn  
**Subject:** Re: Fw:

Dennis --

But if Glen is okay with the narrative, migh we still have a shot at releasing the full data report (not just the results)?  
DanO.

---

**From:** McLerran, Dennis  
**Sent:** Friday, March 01, 2013 9:14:37 AM  
**To:** Eaton, Thomas; Pirzadeh, Michelle; Opalski, Dan  
**Cc:** Hockenbury, Laura; Kelly, Joyce; Stern, Allyn  
**Subject:** Re: Fw:

Thanks everyone on being responsive. Glenn is fine with us releasing just the data next Wednesday foe the FOIA but wants to make sure he reviews the narrative report on the additional data.  
Dennis

---

**From:** Eaton, Thomas  
**Sent:** Friday, March 01, 2013 11:52:40 AM  
**To:** Pirzadeh, Michelle; Opalski, Dan  
**Cc:** Hockenbury, Laura; McLerran, Dennis  
**Subject:** RE: Fw:

I am sending a copy of the Dairy Fed comments, NRCS comments, draft text of the 2/13 Groundwater Report and a copy of our original report to Glen via overnight UPS. It should arrive by tomorrow at 10 AM. I will call Glen and leave him a message to expect it.

---

**From:** Pirzadeh, Michelle  
**Sent:** Thursday, February 28, 2013 7:58 PM  
**To:** Opalski, Dan; Eaton, Thomas  
**Cc:** Hockenbury, Laura  
**Subject:** Fw:

Dan and Tom,

Do either of you have the documents Dennis mentions below readily available? I'll need to try to send hard copies out 1st thing tomorrow morning. Any help would be appreciated. Thanks

Michelle Pirzadeh  
Deputy Regional Administrator  
U.S. Environmental Protection Agency, Region 10  
1200 Sixth Avenue, Suite 900  
Seattle, Washington 98101  
(206) 553-1234

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:57:44 PM  
**To:** Pirzadeh, Michelle  
**Subject:** FW:

Michelle:

Glenn is asking us to Fed Ex him copies of the documents I identify below. He does not have printing capability so he needs hard copies. The address to Fed Ex them to is:

Glenn Paulson

Ex. 6 Personal Privacy (PP)

Ex. 6 Personal Privacy (PP)

If we need to reach him for an address for next week his cell number is Ex. 6 Personal Privacy (PP) Thanks!

Dennis

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:58 PM  
**To:** Paulson, Glenn  
**Cc:** Sussman, Bob; Pirzadeh, Michelle  
**Subject:**

Glenn:

Thanks for talking with me this afternoon. I am confirming that we agreed that a release of the additional well data to the attorney for the citizens' group next Wednesday is OK with you. I will also Fed Ex you a copy of the narrative drafted for the additional data, a copy of the original narrative, a copy of the NRCS comment letter and a copy of the Dairy Federation comment letter.

Since we spoke, I have learned that the attorney for the citizens' group has not agreed to hold off on filing a complaint regarding his FOIA request on the additional well drilling data. So, we will be responding to the press if he goes public with a press release. We have triggered a 10 day extension under the FOIA regulations as we complete our review of the additional data. I still believe it will be appropriate to just release the data to him next Wednesday and follow later with the narrative if needed.

*Dennis J. McLerran  
Regional Administrator  
U.S. EPA, Region 10*

*Office: 206-553-1234  
Fax: 206-553-1809*

Message

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 2/20/2013 3:49:28 AM  
**To:** Dunbar, Bill [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

I think Sussman and Cynthia are going to have to work through that.  
I'll raise it to Bob.  
Dennis

---

**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 10:43:40 PM  
**To:** McLerran, Dennis  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Dennis [Redacted] **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5**

Bill Dunbar  
U.S.E.P.A.

---

**From:** McLerran, Dennis  
**Sent:** Tuesday, February 19, 2013 8:47:42 PM  
**To:** Dunbar, Bill  
**Cc:** Eaton, Thomas; Holsman, Marianne; Pirzadeh, Michelle; Kowalski, Ed; Opalski, Dan; Kelly, Joyce  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Bill:

This looks good [Redacted] **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5** I will forward with a note to Bob S and Jim O'Hara. Thanks!

Dennis

---

**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 8:12:55 PM  
**To:** McLerran, Dennis  
**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

I've added in bold the changes Bob is looking for [Redacted] **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5**

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**From:** Sussman, Bob  
**Sent:** Tuesday, February 19, 2013 1:46 PM  
**To:** McLerran, Dennis; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner,



Nancy

**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

Dennis – this is a good first cut. I would shorten the review of the history (which NRCS should be familiar with) so we can get to the key points in the last half of the letter. I would also put some positive statements upfront along the lines of valuing our relationship with NRCS, wanted to collaborate etc. We might add that we appreciated getting their comments on the report and are considering them very carefully.

And of course the settlement agreement needs to be reflected in the draft.

Once we have a new version, we should probably share with USDA here in DC.

Thanks – BOB

---

**From:** McLerran, Dennis

**Sent:** Thursday, February 14, 2013 7:40 PM

**To:** Sussman, Bob; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce

**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

<< File: Yakima ltr to NRCS 2-14-13ver3.docx >>

Bob and Jim:

Here is a first cut at a letter from me to Washington NRCS Director Roylene Rides-at-the-Door.

Dennis

Message

---

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 2/20/2013 12:20:54 PM  
**To:** Sussman, Bob [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=e85454566bd04951a8b08dcc9df57b4c-Sussman, Bob]  
**Subject:** Fw: Draft Letter Re: Yakima Valley to Washington NRCS  
**Attachments:** Yakima ltr to NRCS 2-19.docx

Bob and Jim:

Attached is a revised draft letter to NRCS.

**Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5**

Dennis

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**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 8:12:55 PM  
**To:** McLerran, Dennis  
**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

I've added in bold the changes Bob is looking for.

**Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5**

---

**From:** Sussman, Bob  
**Sent:** Tuesday, February 19, 2013 1:46 PM  
**To:** McLerran, Dennis; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner, Nancy  
**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

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**From:** McLerran, Dennis  
**Sent:** Thursday, February 14, 2013 7:40 PM  
**To:** Sussman, Bob; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce  
**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

<< File: Yakima ltr to NRCS 2-14-13ver3.docx >>

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Dennis

Message

---

**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 2/21/2013 8:59:58 PM  
**To:** O'Hara.Jim@epa.gov  
**Subject:** FW: NRCS letter  
**Attachments:** Letter to NRCS 2-21.docx

Dennis

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 21, 2013 12:57 PM  
**To:** Sussman, Bob; James O'Hara  
**Cc:** Stoner, Nancy; Giles-AA, Cynthia; Chester, Steven; Holsman, Marianne; Dunbar, Bill; Pirzadeh, Michelle; Opalski, Dan; Eaton, Thomas; Kowalski, Ed; Stern, Allyn; Kelly, Joyce  
**Subject:** FW: NRCS letter

Bob:

Attached is the latest revision to the draft letter to NRCS Washington Director Roylene Rides-at-the-Door. I have tried to shorten it some and have worked on the sentence near the end related to circumstances that might be occurring elsewhere.

**Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5** fully expect it will be a public document shared by the NRCS with dairymen and others. It will also likely be the first point at which it becomes public that we have reached an agreement in principle with the Yakima dairies. Because of that, we should have a communication strategy in place for how this is released and have NRCS share in that along with our Washington State and dairy participants in all of this.

You, Jim O'Hara, Marianne Holsman and I should discuss how we plan to release this and the timing on that in relation to the announcement of the agreement with the dairies.

**Deliberative Process / Ex. 5**

Dennis

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**From:** Dunbar, Bill  
**Sent:** Thursday, February 21, 2013 12:33 PM  
**To:** McLerran, Dennis  
**Subject:** NRCS letter

Message

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**From:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0ACF3804188E4D519D59D67B82EDA2EF-MCLERRAN, DENNIS J.]  
**Sent:** 3/5/2013 4:15:36 PM  
**To:** Paulson, Glenn [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=86a3df55998a4f2ea36893a7a1cce9e1-Paulson, Glenn]  
**CC:** Holsman, Marianne [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Sussman, Bob [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=e85454566bd04951a8b08dcc9df57b4c-Sussman, Bob]; Kelly, Joyce [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=b1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Stern, Allyn [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2e2d6f5df08d4e9abbff2c0880407775-Stern, Allyn]  
**Subject:** Yakima Valley Ground Water Data

Glenn:

I hope your out of office time is going well. As we discussed last Thursday, we are planning to release the additional well drilling data under the FOIA request from CARE attorney Charlie Tebbetts tomorrow so I wanted to check in with you to see how your review of the narrative and the data is going. We have the option of only releasing the data to Mr. Tebbetts tomorrow if you have concerns with the short narrative report. However, if you feel the narrative is ready we could release both the data tables and the narrative tomorrow. As we discussed in our telephone call, we are planning to announce our agreement with four of the dairies in the dairy cluster tomorrow so I would like to hear back from you today if at all possible. If you need to talk with me please call (206) 553-1234 and I will make sure our receptionist finds me so we can talk. Thank you so much for taking the time to review this while you are out of the office. We appreciate it very much.

*Dennis J. McLerran  
Regional Administrator  
U.S. EPA, Region 10*

*Office: 206-553-1234  
Fax: 206-553-1809*

Message

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**From:** Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10FA13022096442D83A97A2B7A699F06-EATON, THOMAS]  
**Sent:** 2/8/2013 12:57:38 AM  
**To:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**CC:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]; Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kelly, Joyce [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=B1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]  
**Subject:** Info for your meeting tomorrow with the NRCS chief  
**Attachments:** Review\_Tarkalson.docx

Hi Dennis,

I did connect with Larry Johnson, NRCS State Engineer this morning. Larry works for Roylene and was one of the two signers of the cover letter transmitting NRCS comments. The other person was Bonda Habits. I didn't get much info from Larry other than he knew about the meeting and had sent some briefing material up to the chief. I asked if he provided any recommendations he could share and he said no recommendations were made.

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After all the groundwater data was collected, Marie's staff contacted NRCS, the Department of Agriculture and the local Conservation District to see if they had any specific information on the dairies in our report that they could share with us. Since most of the information is kept confidential by those agencies, not much specific information was gathered.

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#### **USDA Participation as a peer reviewer**

##### **David Tarkalson**

**Education:** PhD, North Carolina State, Soil Science

**Current Employer:** USDA, Agricultural Research Service, Kimberly Idaho

**Job Title:** Systems agronomist/soil scientist

**Research interests:** Nutrient management, crop production, and water quality and quantity.

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"I have finished my review. Overall the report is well written. The main body of the report is clear and concise. I only have a few suggestions and comments (see attached). Good job."

**Comment by Dr. Tarkalson received during public input:** : Dr. Tarkalson requested that his name and affiliation be

removed as a reviewer of the final report. He stated that the final version of the report contains information that he did not review in the original draft. He cited a couple examples including: 1. inclusion of quantities of lagoon liquid leakage and 2. differing interpretation of pharmaceutical data.

**Response to Dr. Tarkalson:** The conclusions in the draft report that Dr. Tarkalson reviewed indicated dairies were a likely source of nitrates, which we retained in the September report. We did add some additional information on lagoons based on comments from people that we needed more information on the operations of the dairies. We did have the information on pharmaceuticals in the draft report.

Dr. Tarkalson's comments are attached.



Review\_Tarkalso...

Tom Eaton  
Director  
Washington Operations Office  
USEPA, Region 10  
360-753-8086

## Questions

1. Are the purpose, scope, and objectives of the project clear?

**Response:** The purpose of the study is clearly stated at the beginning of the document (1st paragraph in the Introduction). The placement of the purpose at this location is vital to making the document clear to the reader. The scope of the study is also presented clearly in the first paragraph.

2. Is it clear why we selected certain chemical classes (e.g., hormones) or analytical techniques (e.g., isotopic analysis) to serve as potential tracers for nitrate contamination?

**Response:** Overall your explanation of chemical classes is clear. The list of hormones in Table 4 indicates which animals the hormones are used for. There is no mention of any being used in dairy cows. Is this correct? The first 5 listed under “Analyzed at UNL Only” do not indicate for which animals they are from. I am assuming they are associated with mammals in general.

It would be valuable to have some information on the mobility and decay rates of the hormones and pesticides in soils. If an organic constituent is not mobile it is unlikely that they will move to ground water, thus not serve as good indicators of nitrate sources. Need to make sure the selected organic constituents have a purpose not just “this is what the lab can analyze for so we will include” justification behind selection of constituents.

3. Is the experimental design clear?

**Response:** In the main text the design is clear but a few more specifics could be included. These suggestions are listed in other responses.

Regarding Appendix C, some terms need to be better defined – qualified and out of control. When I read “qualified estimated” I think a “scientific guess”. Guessing has no place in scientific investigations. I am assuming that this is not the case, so a better explanation of what “qualified estimates” are would be good. References to QA procedures from other reports are plentiful in this Appendix. Many readers will not search for these so it may be good to summarize them in this report.

After reading Appendix C I had the impression there were a lot of issues with organic compound sample analysis that made me question the validity and need of the data.

4. Is the approach taken for evaluating the isotopic data reasonable given the results from the study and the literature on isotopic analysis (e.g.,  $\delta^{15}\text{N}$ - $\text{NO}_3$  water well values greater than 8.4‰ characterized as dominated by animal waste;  $\delta^{15}\text{N}$ - $\text{NO}_3$  water well values less than 2.0‰



characterized as dominated by fertilizer; and  $\delta^{15}\text{N}$ - $\text{NO}_3$  water wells values between 2.0‰ and 8.4‰ being characterized as isotopically in determinant as to animal waste and/or fertilizer).

**Response:** I am not an expert on isotope analysis but based on what I do know, your approach seems reasonable. Listing the shortcoming of the analysis and what conclusions can be drawn was a good strategy. I would recommend placing the information found in the footnotes of Table 8 into the design section for “D. Isotopic Analysis” on page 27.

5. Are the conclusions supported by the results?

**Response:** The conclusions are supported by the data.

6. Are there results which could be more strongly used to link nitrate contamination to sources?

7. **Response:** Not that I could see.

8. Are the uncertainties adequately addressed and clearly articulated?

9. **Response:** Yes.

#### **Other Comments:**

When giving nitrate concentrations make sure to specify wheather the concentrartios are reported as  $\text{NO}_3$  or  $\text{NO}_3\text{-N}$  (e.g. Background section, Page 5: “...0.5 mg/L and to 1.1 mg/L...”)

In Figure 1, some leaching path arrows look line runoff paths.

Including Section X. Study Limitations and Uncertainties was important. Regarding the second paragraph discussion on samples from wells with high nitrate and low oxygen, denitrification will not proceed under these situation unless there is an energy source (usually organic C) for the denitrifying bacteria to use and a terminal electron acceptor ( $\text{NO}_3^-$  in this case due to lack of a better electron acceptor,  $\text{O}_2$ ). These waters could have been low in organic C thus accounting for the high levels of nitrate with low oxygen in the water. Did you measure organic C content of the water? This point (requirement of an energy source) was missing in the introduction section dealing with denitrification.

Message

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**From:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133B66D6AB1B42108751E37B28176EC3-DUNBAR, BILL (WILLIAM) D.]  
**Sent:** 2/8/2013 1:23:39 AM  
**To:** McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**CC:** Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kelly, Joyce [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=B1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]  
**Subject:** Re: Info for your meeting tomorrow with the NRCS chief  
**Attachments:** NRCS WA cvr ltr and 3 attmnts.pdf

Dennis - I'm sure you'll hear about it from them, so it's also important to know that NRCS has provided three separate critiques of our study. all three are in the PDF attached here.

--Bill.



NRCS WA cvr ltr  
and 3 attmnts.pdf

**Bill Dunbar**

Policy Advisor

U.S. Environmental Protection Agency, Region 10

1200 Sixth Avenue, Suite 900

Mail Stop RA-140

Seattle, WA 98101

206/553-1019

Cell: 206/245-7452

**From:** Thomas Eaton/R10/USEPA/US  
**To:** Dennis McLerran/R10/USEPA/US@EPA  
**Cc:** Bill Dunbar/R10/USEPA/US@EPA, Dan Opalski/R10/USEPA/US@EPA, Joyce Kelly/R10/USEPA/US@EPA  
**Date:** 02/07/2013 04:57 PM  
**Subject:** Info for your meeting tomorrow with the NRCS chief

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[attachment "Review\_Tarkalson.docx" deleted by Bill Dunbar/R10/USEPA/US]

Tom Eaton  
Director  
Washington Operations Office  
USEPA, Region 10  
360-753-8086



Natural Resources Conservation Service  
316 W. Boone Ave. Suite 450  
Spokane, WA 99201-2348

#45

phone 509-323-2900  
fax 509-323-2909  
web site [www.wa.nrcs.usda.gov](http://www.wa.nrcs.usda.gov)

#28.1  
28.2

December 7, 2012

Office of Environmental Assessment (OEA)  
Attn: Carol Harrison  
U.S. EPA, Region 10  
1200 Sixth Avenue, Suite 900  
Mail code: OEA-095  
Seattle, WA 98101

**RE: NRCS Comments - Lower Yakima Valley Nitrate Study**

Dear Ms. Harrison:

The Natural Resources Conservation Service (NRCS) is submitting comments for consideration regarding the Lower Yakima Valley Nitrate Study.

NRCS national discipline specialists for nutrient management and agricultural waste management have reviewed the study report and provided comments regarding the methodologies, procedures and findings in the study. Their comments are provided in attachments two and three.

In addition to our national discipline specialists comments, state staff have provide a few additional comments which are provided in attachment one.

NRCS hopes that these comments are helpful in finalizing the study. Please do not hesitate to call if you have any question.

Sincerely,

Bonda Habets  
State Resource Conservationist

Lawrence A. Johnson, P.E.  
State Conservation Engineer

Attachments (3)

cc: Roylene Rides-at-the-Door, State Conservationist

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## ATTACHMENT 1

### Washington State NRCS comments:

The study does not accurately represent the likely discharge rate occurring from Dairy Waste Storage Ponds (WSP) identified in the report. The assessment utilized by EPA was based on limited site information. Assumed values were utilized for computing the specific discharge, as such the results and findings are erroneous and unreliable. We recommend that EPA take immediate steps to revise the findings taking into consideration the following:

1. Accurate estimates for computing the likely discharge amounts require site specific information of the WSP. The WSP wetted surface soil type and extent is required. If the WSP is lined the liner thickness is required and the seasonal fluctuation of the effluent level acting on the WSP wetted surface must be known. Utilizing the design guidance found in the NRCS Agricultural Waste Management Field Handbook (AWFMH), Appendix 10D, for computing the specific discharge rate from a WSP requires very detailed data. Given that the actual WSP data is not available for any of these calculations, all the estimates for specific discharge rates are erroneous and unreliable.
2. NRCS design methodology for computing WSP specific discharge rates are based on very accurate permeability rate determinations. The procedure for conducting the analysis is found in ASTM D-5084, "Standard test methods for Measurement of Hydraulic Conductivity of Saturated porous Materials using a Flexible Wall Permeameter". EPA should collect multiple undisturbed soils samples from each WSP within the wetted surface area that would be representative of the entire wetted surface of the WSP. The results should be used to revise the seepage rates based on the procedure in the AWFMH, Appendix 10D.
3. The EPA study assumption that the WSP is full year round is invalid for estimating and quantifying the discharge occurring from a WSP. The methodology should be revised to estimate the discharge over a full year as the WSP is being filled and then emptied. To assume a full WSP condition for specific discharge quantification is unrealistic and unfairly overestimates the discharge.
4. The maximum effluent depth over the liner is required for estimating specific discharge rates. EPA should conduct topographical surveys of each evaluated WSP to determine the WSP depth and effluent levels acting on the wetted surface area of the WSP.
5. NRCS recommends that greater consideration and focus be given to other nitrate sources that have a higher potential to impact groundwater than the WSP's. The organic and synthetic material used for fertilizer across all the crop acres if not applied at the right amount, the right time and the right place has a higher chance of impacting the groundwater quality than the WSP specified. NRCS is well equipped to provide technical and financial assistance for the development and implementation of a comprehensive nutrient management plan.
6. The EPA study mentioning a difference in human waste being treated and animal waste is not, is misleading in that treated human waste (biosolids) still poses a great threat for contamination of nitrates and heavy metals when discharged into the environment. *J. where*
7. The EPA study claiming the amount of waste produced by the cluster was similar to a community of more than 2,827,000, dismissed the fact that 65% of the study area is land application, where the majority of the animal waste is utilized by crops with high nitrogen needs. Only some of the fields were noted in WSDA inspector reports to have elevated levels of nitrogen.

## ATTACHMENT 2

### ***Yakima Valley Nitrate Study***

#### **Comments/Input**

The following is being provided in response to the USEPA report, EPA-910-R-12-003, entitled *Relation between Nitrate in Water Wells and Potential Sources in the Lower Yakima Valley, Washington*.

**Introduction.** This EPA study was generated primarily due to the concerns by many pertaining to the high levels of nitrate in water wells in the Lower Yakima Valley, WA. In an effort to determine the source of those high nitrates, dairy farms (along with other minor sources) located within proximity to the wells, were identified as potential sources, specifically dairy farms in operation there. The report concludes that dairy producer's facilities and their associated lands are the principal sources of high nitrates in the wells. To validate their claim, EPA collected much data and performed numerous tests attempting to pinpoint the source of the nitrates.

After reading the report and reviewing the numerous data and conclusions derived from it, I am highly skeptical of their conclusions based on their testing methods, procedures, and interpretations of these recorded throughout the report. The report is filled with considerable errors in calculations and interpretations that, based on the performed tests, methods and interpretations, cannot be scientifically defended to derive at the stated conclusions. I want to acknowledge that high nitrate levels in water wells are a documented health hazard. However, the source and extent of those nitrates must be accurately identified before corrective measures can be implemented or a course of action, pineal or otherwise, is undertaken.

**Comments/Input.** The study design itself is flawed in that it fully acknowledges that identification of the extent and sources of nitrates did not take into account any losses of nitrates from biological, physical, or chemical processes and also did not account for crop utilization (ref. page 16, Phase 3 Study Results). The conclusions pertaining to the dairies are based on data that does not properly represent the sources or the extent of nitrates. In essence, the study simply collects data on particular sites at particular times and finds nitrogen of various forms and concludes that all these are the cause of the well water contamination or substantially contribute to it.

Anytime animal wastes and fertilizers are utilized to provide the required nutrients for crops and forages, a minimum set of information must be considered to properly provide a management system that simultaneously provides needed plant nutrients as well as protects surface and subsurface waters. To accurately determine what the needs of the crop are consideration *must* be given as to the proper rate, proper timing, the proper source, and the proper place of nutrient applications including fertilizers and manures. Lack of consideration to any of these will lead to potential production and or environmental problems. Additionally, ignoring any of these while diagnosing an environmental or production problem will also result in less than accurate conclusions. In attempting to determine the source and extent of the nitrate problem in the Lower Yakima Valley, this report ignored or misinterpreted vital data and information to derive at their conclusions.

1. The report does not utilize actual design parameters of the waste storage pond for any of their calculations for all the estimates of specific discharge. The only dimension that was measured is the top surface area of the waste storage pond. The size of the top of this pond was measured at 6.175 acres, which is a significant structure.

By NRCS design criteria, written in the Waste Storage Facility, CPS 313 (2004), the storage ponds would have a designed specific discharge as little as 0.07 inches and is equivalent less than 1% of the total annual depth of precipitation in this part of the state.

Given that the basis of concern stated in the EPA report is groundwater quality, there conclusions that the ponds are causing considerable leaching of nitrates into the groundwater is incorrect. Proper calculations would conclude that the ponds are **not** likely the source of nitrates in the wells. This also would suggest that there may be other nitrate sources that have a higher potential to impact groundwater than the waste storage ponds as stated.

2. The EPA report suggests that irrigated cropland is expected to be a likely source of nitrates in drinking water wells (ref. page ES-9). However, the only pathway that nitrate contaminants can enter well water is through either:
  - 1). Leaching through the soil profile past the crop/forage rooting zone into ground water and subsequent movement underground to a well, or
  - 2). through surface water flow off of the field and directly into a recharge area that feeds the well itself.

In consideration of leaching, the report attempts to identify that the soils on a majority of the fields that are receiving manures and commercial fertilizer are considered "well drained" and that they have "saturated

hydraulic conductivity” characteristics that is considered high (ref. EPA-910-R-12-003, surface soils, page 35 and Appendix B). The report cites the USDA NRCS soil survey and reports generated from the survey to characterize the fields.

Saturated Hydraulic Conductivity. According to the definition that USDA NRCS uses to describe and characterize saturated hydraulic conductivity (Soil Survey Manual, Ag Handbook 18) saturated flow occurs only when the soil water pressure is positive; that is, when the soil matric potential is zero (satiated wet condition). This situation takes place when about 95% of the total pore space is filled with water (5% is air). If the soil remains saturated for a prolonged period (several months or longer) the percent of total pore space filled with water may approach 100 percent. “Saturated hydraulic conductivity CANNOT be used to describe water movement under unsaturated conditions” (ref. USDA Soil Survey Manual, Soil Survey Division Staff, Agriculture handbook 18, October 1993, page 103).

Because irrigation water management details were not collected from the producers nor was data collected in the field that measured soil saturation or duration, there is no data that can substantiate that the simple classification of hydraulic conductivity precludes leaching. Therefore, the data that EPA draws upon in this report to suggest the fields were leaching is circumstantial and cannot be used to conclude that leaching is attributed to the land treatment fields where manures and fertilizers were applied.

Drainage Class. Natural drainage class refers to the frequency and duration of wet periods under conditions similar to those under which the soil developed. Alterations of the water regime by man, either drainage or irrigation, is NOT considered unless the alterations have significantly changed the morphology of the soil (ref. USDA Soil Survey Manual, Soil Survey Division Staff, Agriculture handbook 18, October 1993, page 98).

The USDA Soil Survey manual describes “well drained” as *water is removed from the soil readily but not rapidly. Internal free water occurrence commonly is deep or very deep; annual duration is not specified. Water is available to plants throughout most of the growing season in humid regions. Wetness does not inhibit growth of roots for significant periods during most of the growing seasons* (ref. USDA Soil Survey Manual, Soil Survey Division Staff, Agriculture handbook 18, October 1993, page 98).

The EPA report misinterprets the definition of “well drained” tying nitrate leaching to the natural drainage classification of the soil. Again, there is no documentation of any kind that would lead to the conclusion that, based on the natural drainage classification of a soil, leaching or subsurface water contamination occurred in the Lower Yakima Valley.

3. The report makes substantial conclusions pertaining to the potential for nitrate leaching and runoff based on soil tests that were derived from the top 1 inch of soil (Table ES-1, footnote b). The top 1-3 inches of soil contains a large majority of the soil profile’s organic matter, where large amounts of organic matter and mineralization occur. Additionally, depending upon when soil samples were collected and how and when any manures or fertilizers were applied, it is not uncommon to see high quantities of N near the soil surface for certain periods of the year. For example, if manure was broadcast on the soil surface and not incorporated, higher quantities of N-P-and K would be apparent until such time as the manures or fertilizers were incorporated or volatilized. Management techniques have a great deal to do with the location and quantities of nutrients in the soil profile depending upon the characteristics of the nutrient and the management practices utilized.

To accurately ascertain if nitrates were moving through the soil profile, deep soil tests (36-60 inches) should have been collected from fields above gradient and below gradient of affected wells. If nitrates were found below the rooting zone of the crops grown, this may have been an indicator of potential nitrate movement to ground water. A 1 inch soil sample has limited applicability if any.

4. Some of the testing interpretations are also questionable as to their applicability. For example, on page 20 of the report, EPA states that they tested for nitrate, nitrite, ammonia, and TKN. “Total nitrogen concentration was calculated by summing concentration of nitrate, nitrite, and TKN”. The TKN test is used to determine what the potential total of various forms of N are. A TKN test is the measure of organic N (nitrate and nitrite), ammonia N (NH<sub>3</sub>) and ammonium N (NH<sub>4</sub>). By adding nitrite and nitrate to the TKN, you essentially are adding quantities of nitrate and nitrite twice, substantially increasing the total. Nitrate and nitrite are ion specific. Additionally, TKN tests include N that has not been mineralized (as nitrates) and assumptions that they will be are erroneous. The nitrogen cycle, as shown in the report, includes mineralization, denitrification, volatilization, etc. Not all nitrite will be nitrate. Not all nitrate will remain nitrate (denitrification) especially under wet or saturated soil conditions. Summarily, the results of the improper (depth) soil test data is being misinterpreted and cannot be used to draw conclusion as to the source of nitrates in the wells.

**Summary.** Due to the fact that specific data pertaining to crop management and tillage systems, manure management, irrigation water management, nutrient management, and pest management were not collected or were not available to EPA, utilizing gross or generalized characteristics or data pertaining to soils within the Lower Yakima Valley for purposes of identifying sources of nitrates in well water is not accurate or conclusive.

It is my suggestion that this report be retracted and data collection begin in earnest including the above listed management information and the appropriate tests using proper data collection methods and testing techniques. The results of which should be independently analyzed by non-affected parties to enable proper conclusions as to the source of nitrates in water wells in the Lower Yakima Valley.

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### ATTACHMENT 3

Thank you for the opportunity to provide comments on the EPA- Relation Between Nitrate in Water Wells and Potential Sources in the Lower Yakima Valley, Washington study.

At the request of NRCS Washington State leadership, NRCS National Science and Technology staff has reviewed the report and submits the following comments and observations:

- 1) The use of generalized recommendations contained in the NRCS Animal Waste Management Field Handbook (AWMFH) to predict the seepage loss contribution from individual farms is misleading. AWMFH recommendations and data are well supported by research and field experience throughout the US. However, they are generalized recommendations that typically get adjusted to accommodate site conditions during the planning and/or installation phases of storage facility construction. Site specific testing and on-site evaluations are usually necessary to assure adequate design for a given site. Planning for an adequate structure requires detailed information about anticipated manure volumes, manure type and consistency, climate, and production area runoff, etc.
- 2) The generalized AWMFH seepage rates should not be used to predict leakage losses from multiple dairies distributed randomly in a major watershed. Generalized seepage rates are used by planners/installers to anticipate the potential for loss and the need for additional conservation practices that will provide adequate protection of local water quality. Recent Mississippi River Basin and Chesapeake Bay Watershed CEAP studies have established the effectiveness of installed NRCS conservation practices at protecting water quality. Practices were most effective when they were coordinated as a suite of practices designed to deal with a site-specific water quality issue.
- 3) NRCS funded waste storage structures must be constructed following engineering conservation practice standards that are maintained by state-based NRCS technical experts. To receive USDA funding, producers must sign a contract that requires compliance with design, oversight/maintenance, and other criteria established by the practice standard. The contract is permanently tied to the state's official practice standard when the contract was signed by the producer, i.e., lagoons built 15-20 years ago are not associated with contracts that enforce criteria for the current official practice standard.
- 4) EPA's restricted access to important sites or data made it difficult to estimate the pollution contribution from individual farms. Farm specific information would have helped the researchers isolate site/management issues contributing contaminants to the environment.
- 5) The use of aerial photography to determine storage volumes can grossly over estimate each operation's manure storage capacity and therefore your estimate of seepage losses.
- 6) A storage facility is typically designed following rigid engineering standards, and local code usually requires routine monitoring for failure. EPA does not know if the structures were designed per code or if competent engineers were involved in the work. There are many factors that could lead to a leaking storage facility. Targeting all operations upgrade of a contaminated well may unfairly focus attention on well managed operations.
- 7) EPA does not know if the operations have storage facilities that were designed to handle the number of animals confined. If not, the lagoons may lack sufficient capacity to accommodate the manure/wastewater volumes generated, making them vulnerable to discharge. Has a nutrient management plan been followed to assure adequate facilities to handle mortality, spoiled feed, contaminated runoff, medical wastes, etc? Are animals fenced out of streams; are nutrient being applied too near unbuffered streams? These factors were not considered by the study.
- 8) Every AFO deals with a unique set of circumstances that define how to best manage large volumes of potentially polluting materials. Site factors, e.g., soil, topography, climate, animal type/number, period of confinement, confinement facility type, field hydrology, storage design/volume, spreadable acres available, and management styles change significantly from farm to farm. A leaking lagoon may not always be the principal problem, and oftentimes additional conservation practices, or better management of installed practices, may significantly reduce losses.

- 9) It appears that the high levels of nitrate found in drinking water wells in the Yakima Valley are coming from multiple sources. More information is needed to help focus attention on site/management issues contributing pollutants.
- 10) NRCS acknowledges the need to encourage producers to install sufficient conservation measures to minimize the movement of potential contaminants off-site. Accordingly, NRCS manages approximately 190 conservation standards that can be used to protect air, soil and water quality. Structural practices are available to help the producer keep air and water clean in the confinement (production) area, and land treatment practices (cropland, hayland, pasture) help growers minimize non-point source losses associated with erosion, leaching, volatilization, denitrification and surface flow.
- 11) Livestock growers enrolled in USDA programs are strongly encouraged to manage their manure handling, storage and field allocation activities following a Comprehensive Nutrient Management Plan (CNMP). The CNMP is developed based on site conditions and is designed to help farmers safely apply stored manure to their available land base.
- 12) The NRCS Nutrient Management Conservation Practice Standard (CPS 590) provides the minimum nutrient management planning criteria for the application of nutrients (synthetic or organic) to agricultural lands enrolled in USDA programs.
- 13) In January 2012, NRCS released a revised nutrient management policy and CPS 590. The new policy encourages improved nitrogen and phosphorus risk assessment tools, precision and enhanced efficiency fertilizer technologies, suites of coordinated conservation practices, and adaptive nutrient management strategies. Improved nutrient use efficiency saves the producer money and also reduces the potential for loss to the environment.
- 14) NRCS is committed to working with farmers to help them minimize the impact that farming operations can have on local water quality, including drinking water.

Message

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**From:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133B66D6AB1B42108751E37B28176EC3-DUNBAR, BILL (WILLIAM) D.]  
**Sent:** 2/20/2013 1:12:55 AM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**CC:** Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS  
**Attachments:** Yakima ltr to NRCS 2-19.docx

I've added in bold the changes Bob is looking for.

**Deliberative Process / Ex. 5**

## Deliberative Process / Ex. 5



Yakima ltr to  
NRCS 2-19.docx

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**From:** Sussman, Bob  
**Sent:** Tuesday, February 19, 2013 1:46 PM  
**To:** McLerran, Dennis; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner, Nancy  
**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

Dennis -- this is a good first cut. I would shorten the review of the history (which NRCS should be familiar with) so we can get to the key points in the last half of the letter. I would also put some positive statements upfront along the lines of valuing our relationship with NRCS, wanted to collaborate etc. We might add that we appreciated getting their comments on the report and are considering them very carefully.

And of course the settlement agreement needs to be reflected in the draft.

Once we have a new version, we should probably share with USDA here in DC.

Thanks -- BOB

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 14, 2013 7:40 PM  
**To:** Sussman, Bob; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce  
**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

<< File: Yakima ltr to NRCS 2-14-13ver3.docx >>

Bob and Jim:

Here is a first cut at a letter from me to Washington NRCS Director Roylene Rides-at-the-Door.

Dennis

Message

**From:** Kowalski, Ed [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=05111EC2F4314434A376F1C84C851820-KOWALSKI, ED]  
**Sent:** 2/20/2013 3:25:49 AM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]; Dunbar, Bill [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]  
**CC:** Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Holsman, Marianne [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]; Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kelly, Joyce [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=b1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Bill, not seeing the latest iteration, can I assume you will add the Cynthia Giles language Tom raised at today's ET meeting?

---

**From:** McLerran, Dennis  
**Sent:** Tuesday, February 19, 2013 5:47:42 PM  
**To:** Dunbar, Bill  
**Cc:** Eaton, Thomas; Holsman, Marianne; Pirzadeh, Michelle; Kowalski, Ed; Opalski, Dan; Kelly, Joyce  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Bill:

This looks good **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5** and I will forward with a note to Bob S and Jim O'Hara. Thanks!

Dennis

---

**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 8:12:55 PM  
**To:** McLerran, Dennis  
**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

I've added in bold the changes Bob is looking for **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5**

---

**From:** Sussman, Bob  
**Sent:** Tuesday, February 19, 2013 1:46 PM  
**To:** McLerran, Dennis; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner,

Nancy

**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

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Message

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**Sent:** 2/20/2013 3:43:40 AM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Dennis: **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5** Ed mentioned today that that is a particularly important notion to include.

Bill Dunbar  
U.S.E.P.A.

---

**From:** McLerran, Dennis  
**Sent:** Tuesday, February 19, 2013 8:47:42 PM  
**To:** Dunbar, Bill  
**Cc:** Eaton, Thomas; Holsman, Marianne; Pirzadeh, Michelle; Kowalski, Ed; Opalski, Dan; Kelly, Joyce  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Bill:

This looks good! **Deliberative Process / Ex. 5**

**Deliberative Process / Ex. 5** I will forward with a note to Bob S and Jim O'Hara. Thanks!

Dennis

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**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

I've added in bold the changes Bob is looking for... **Deliberative Process / Ex. 5**  
**Deliberative Process / Ex. 5**

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**Sent:** Tuesday, February 19, 2013 1:46 PM  
**To:** McLerran, Dennis; James O'Hara  
**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner, Nancy  
**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

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**Sent:** Thursday, February 14, 2013 7:40 PM

**To:** Sussman, Bob; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce

**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

<< File: Yakima ltr to NRCS 2-14-13ver3.docx >>

Bob and Jim:

Here is a first cut at a letter from me to Washington NRCS Director Roylene Rides-at-the-Door.

Dennis



Message

---

**From:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133B66D6AB1B42108751E37B28176EC3-DUNBAR, BILL (WILLIAM) D.]  
**Sent:** 2/20/2013 3:50:40 AM  
**To:** Kowalski, Ed [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=05111ec2f4314434a376f1c84c851820-Kowalski, Ed]; McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**CC:** Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Holsman, Marianne [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]; Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kelly, Joyce [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=b1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Did that this afternoon. Dennis has it now.  
Bill Dunbar  
U.S.E.P.A.

---

**From:** Kowalski, Ed  
**Sent:** Tuesday, February 19, 2013 10:25:49 PM  
**To:** McLerran, Dennis; Dunbar, Bill  
**Cc:** Eaton, Thomas; Holsman, Marianne; Pirzadeh, Michelle; Opalski, Dan; Kelly, Joyce  
**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

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**Subject:** Re: Draft Letter Re: Yakima Valley to Washington NRCS

Bill:

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Deliberative Process / Ex. 5

Deliberative Process / Ex. 5 I will forward with a note to Bob S and Jim O'Hara. Thanks!

Dennis

---

**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 8:12:55 PM  
**To:** McLerran, Dennis  
**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

## Deliberative Process / Ex. 5

---

**From:** Sussman, Bob

**Sent:** Tuesday, February 19, 2013 1:46 PM

**To:** McLerran, Dennis; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner, Nancy

**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

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Thanks -- BOB

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**From:** McLerran, Dennis

**Sent:** Thursday, February 14, 2013 7:40 PM

**To:** Sussman, Bob; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce

**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

<< File: Yakima ltr to NRCS 2-14-13ver3.docx >>

Bob and Jim:

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Dennis

Message

---

**From:** Sussman, Bob [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=E85454566BD04951A8B08DCC9DF57B4C-SUSSMAN, BOB]  
**Sent:** 2/20/2013 3:22:09 PM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**Subject:** RE: Draft Letter Re: Yakima Valley to Washington NRCS

We'll need to discuss **Deliberative Process / Ex. 5**

I still think the front part of the letter can be cut back.

---

**From:** McLerran, Dennis  
**Sent:** Wednesday, February 20, 2013 7:21 AM  
**To:** Sussman, Bob  
**Subject:** Fw: Draft Letter Re: Yakima Valley to Washington NRCS

Bob and Jim:

Attached is a revised draft letter to NRCS.

**Deliberative Process / Ex. 5**

## **Deliberative Process / Ex. 5**

Dennis

---

**From:** Dunbar, Bill  
**Sent:** Tuesday, February 19, 2013 8:12:55 PM  
**To:** McLerran, Dennis  
**Cc:** Eaton, Thomas  
**Subject:** FW: Draft Letter Re: Yakima Valley to Washington NRCS

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**Deliberative Process / Ex. 5**

## **Deliberative Process / Ex. 5**

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**Sent:** Tuesday, February 19, 2013 1:46 PM  
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**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce; Gilinsky, Ellen; Stoner, Nancy  
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**To:** Sussman, Bob; James O'Hara

**Cc:** Holsman, Marianne; Dunbar, Bill; Eaton, Thomas; Opalski, Dan; Kowalski, Ed; Kelly, Joyce

**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS

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Dennis

Message

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**From:** Paulson, Glenn [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=86A3DF55998A4F2EA36893A7A1CCE9E1-PAULSON, GLENN]  
**Sent:** 3/1/2013 4:54:36 AM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]  
**CC:** Sussman, Bob [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=e85454566bd04951a8b08dcc9df57b4c-Sussman, Bob]; Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]  
**Subject:** Re:

Noted, this does not change my view on how to proceed, Glenn

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 6:54:37 PM  
**To:** Paulson, Glenn  
**Cc:** Sussman, Bob; Pirzadeh, Michelle  
**Subject:**

Glenn:

Thanks for talking with me this afternoon. I am confirming that we agreed that a release of the additional well data to the attorney for the citizens' group next Wednesday is OK with you. I will also Fed Ex you a copy of the narrative drafted for the additional data, a copy of the original narrative, a copy of the NRCS comment letter and a copy of the Dairy Federation comment letter.

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*Dennis J. McLerran  
Regional Administrator  
U.S. EPA, Region 10*

*Office: 206-553-1234  
Fax: 206-553-1809*

Message

---

**From:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133B66D6AB1B42108751E37B28176EC3-DUNBAR, BILL (WILLIAM) D.]  
**Sent:** 2/13/2013 10:09:52 PM  
**To:** Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]; Holsman, Marianne [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]  
**Subject:** Critical comments from public agencies  
**Attachments:** A sampling of critical comments from public agencies.docx



A sampling of  
critical comment...

**Bill Dunbar**

Policy Advisor  
U.S. Environmental Protection Agency, Region 10  
1200 Sixth Avenue, Suite 900  
Mail Stop RA-140  
Seattle, WA 98101  
206/553-1019  
Cell: 206/245-7452

Message

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**From:** Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10FA13022096442D83A97A2B7A699F06-EATON, THOMAS]  
**Sent:** 2/13/2013 10:42:19 PM  
**To:** Dunbar, Bill [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]  
**CC:** Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; McLerran, Dennis [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]; Holsman, Marianne [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]  
**Subject:** Re: Critical comments from public agencies

Hi Bill,

We are able to respond to several of these comments with logical, defensible answers, the new data addresses others and we may have to make some changes/clarifications to address some.

For example, the criticism about sampling the top 1" of soil is a misunderstanding by soil scientists regarding the purpose of our sampling. We were trying to detect constituents that might be detectable in groundwater such as pesticide residues. We were not trying to use this data to quantify or estimate the amount of nitrates that could leach into the groundwater. For this purpose, I would argue that collecting a 1" soil sample is appropriate.

Since the main job of several of the commenters is to work with farmers to estimate nutrient needs for given crops and to limit commercial fertilizer or manure application to meet those needs, they correctly stated that a 1" soil sample is not appropriate for that purpose. We did not collect it for that purpose and we can respond accordingly.

I am still curious why BIA chose to send in comments, but don't read theirs as that critical

Tom Eaton  
Director  
Washington Operations Office  
USEPA, Region 10  
360-753-8086

**From:** Bill Dunbar/R10/USEPA/US  
**To:** Dan Opalski/R10/USEPA/US@EPA, Thomas Eaton/R10/USEPA/US@EPA, Dennis McLerran/R10/USEPA/US@EPA, Marianne Holsman/R10/USEPA/US@EPA  
**Date:** 02/13/2013 02:09 PM  
**Subject:** Critical comments from public agencies

[attachment "A sampling of critical comments from public agencies.docx" deleted by Thomas Eaton/R10/USEPA/US]

**Bill Dunbar**  
Policy Advisor  
U.S. Environmental Protection Agency, Region 10  
1200 Sixth Avenue, Suite 900  
Mail Stop RA-140  
Seattle, WA 98101  
206/553-1019  
Cell: 206/245-7452

Message

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**From:** Opalski, Dan [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=8B5ED6410D934BF699A008A252791A55-OPALSKI, DAN]  
**Sent:** 3/1/2013 5:30:42 PM  
**To:** McLerran, Dennis [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=0acf3804188e4d519d59d67b82eda2ef-McLerran, Dennis J.]; Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Pirzadeh, Michelle [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2ae309e0a52a42cd847709b39bc2239a-Pirzadeh, Michelle]  
**CC:** Hockenbury, Laura [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=ccf5c33e87eb46168e43d4fe8b5bb9ba-Hockenbury, Laura A.]; Kelly, Joyce [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=b1487930f7634eecbe7c90bbd3003599-Kelly, Joyce]; Stern, Allyn [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=2e2d6f5df08d4e9abbff2c0880407775-Stern, Allyn]  
**Subject:** Re: Fw:

Dennis ---

But if Glen is okay with the narrative, might we still have a shot at releasing the full data report (not just the results)?  
DanO.

---

**From:** McLerran, Dennis  
**Sent:** Friday, March 01, 2013 9:14:37 AM  
**To:** Eaton, Thomas; Pirzadeh, Michelle; Opalski, Dan  
**Cc:** Hockenbury, Laura; Kelly, Joyce; Stern, Allyn  
**Subject:** Re: Fw:

Thanks everyone on being responsive. Glenn is fine with us releasing just the data next Wednesday for the FOIA but wants to make sure he reviews the narrative report on the additional data.  
Dennis

---

**From:** Eaton, Thomas  
**Sent:** Friday, March 01, 2013 11:52:40 AM  
**To:** Pirzadeh, Michelle; Opalski, Dan  
**Cc:** Hockenbury, Laura; McLerran, Dennis  
**Subject:** RE: Fw:

I am sending a copy of the Dairy Fed comments, NRCS comments, draft text of the 2/13 Groundwater Report and a copy of our original report to Glen via overnight UPS. It should arrive by tomorrow at 10 AM. I will call Glen and leave him a message to expect it.

---

**From:** Pirzadeh, Michelle  
**Sent:** Thursday, February 28, 2013 7:58 PM  
**To:** Opalski, Dan; Eaton, Thomas  
**Cc:** Hockenbury, Laura  
**Subject:** Fw:

Dan and Tom,

Do either of you have the documents Dennis mentions below readily available? I'll need to try to send hard copies out 1st thing tomorrow morning. Any help would be appreciated. Thanks



Michelle Pirzadeh  
Deputy Regional Administrator  
U.S. Environmental Protection Agency, Region 10  
1200 Sixth Avenue, Suite 900  
Seattle, Washington 98101  
(206) 553-1234

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:57:44 PM  
**To:** Pirzadeh, Michelle  
**Subject:** FW:

Michelle:

Glenn is asking us to Fed Ex him copies of the documents I identify below. He does not have printing capability so he needs hard copies. The address to Fed Ex them to is:

Glenn Paulson

Ex. 6 Personal Privacy (PP)

Ex. 6 Personal Privacy (PP)

If we need to reach him for an address for next week his cell number is Ex. 6 Personal Privacy (PP) Thanks!

Dennis

---

**From:** McLerran, Dennis  
**Sent:** Thursday, February 28, 2013 5:58 PM  
**To:** Paulson, Glenn  
**Cc:** Sussman, Bob; Pirzadeh, Michelle  
**Subject:**

Glenn:

Thanks for talking with me this afternoon. I am confirming that we agreed that a release of the additional well data to the attorney for the citizens' group next Wednesday is OK with you. I will also Fed Ex you a copy of the narrative drafted for the additional data, a copy of the original narrative, a copy of the NRCS comment letter and a copy of the Dairy Federation comment letter.

Since we spoke, I have learned that the attorney for the citizens' group has not agreed to hold off on filing a complaint regarding his FOIA request on the additional well drilling data. So, we will be responding to the press if he goes public with a press release. We have triggered a 10 day extension under the FOIA regulations as we complete our review of the additional data. I still believe it will be appropriate to just release the data to him next Wednesday and follow later with the narrative if needed.

**Dennis J. McLerran**  
*Regional Administrator*  
*U.S. EPA, Region 10*

*Office: 206-553-1234*  
*Fax: 206-553-1809*

Message

---

**From:** Winiecki, Eric [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=45FA974E305B49CD8AF513001A800543-WINIECKI, ERIC]  
**Sent:** 3/4/2013 6:22:30 PM  
**To:** Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; Cox, Michael [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=Cddd6a5bb3c2477183799ef56cdb464f-Cox, Michael]; MacDonald, Jennifer [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=507c2f57e60741b2b137cdf441c1c731-MacDonald Jennifer]; Fleming, Sheila [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=5d3e29dad0ec4cd98d87e1a142435c57-Fleming, Sheila]; Fuentes, Rene [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=3eb2a2e594934952b91ca7ade37262c5-Ruentes, Rene]; Jennings, Marie [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=Ec4bd403e1994b80976542bb9458ae72-Jennings, Marie]  
**Subject:** RE: Updated Lagoon Section  
**Attachments:** Yakima Comments Lagoon March 4 2013.docx; Lagoon-NRCSCriteriaRevisions.pdf

Thanks Tom,

I've incorporated your changes and others received, and added a new sentence about the Ecology study.

There is a remaining question about the meaning of a "65%" number in one of the comments toward the end, but I think overall the lagoon section's in pretty good shape.

Eric

---

**From:** Eaton, Thomas  
**Sent:** Monday, March 04, 2013 8:22 AM  
**To:** Cox, Michael; Winiecki, Eric; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie  
**Subject:** RE: Updated Lagoon Section

Looks good - a few minor edits

---

**From:** Cox, Michael  
**Sent:** Friday, March 01, 2013 4:54 PM  
**To:** Winiecki, Eric; Eaton, Thomas; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie  
**Cc:** Cox, Michael  
**Subject:** RE: Updated Lagoon Section

If you want to comment on the lagoon section please use this version. It is in the right format and will make easier to insert. I combined a few of the comments and responses but no change in the content.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
[cox.michael@epa.gov](mailto:cox.michael@epa.gov)

---

**From:** Winiecki, Eric  
**Sent:** Friday, March 01, 2013 4:10 PM

**To:** Cox, Michael; Eaton, Thomas; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie

**Subject:** Updated Lagoon Section

Hi folks,

I incorporated Tom's comments into the lagoon responses and made some additional improvements. Please review this version. The second file above is an attachment that is referenced in one of the responses.

Thanks,

Eric

## **Appendix 2**

### **WSP Practice Standard Criteria Reference Tools**

#### **Table outline for – NRCS Practice Standard Criteria Revisions**

**Waste Storage Pond, PS-425**

**Dated: 1979-1994**

**Waste Storage Facility, PS-313 (Includes Pond Criteria)**

**Dated 2000- Current**

#### **313 PRACTICE STANDARD PERFORMANCE MEASURE CHECKLIST**

#### **Washington State NRCS REVISION Dates:**

- April 1979
- February 1987
- January 1994
- February 2000
- June 2001
- December 2004

<b>Common pond construction dimension criteria for all WSP practices and all revisions: April 1979 to December 2004</b>		
<b>Minimum Top Width</b>	<b>Inside and Outside Slopes</b>	<b>Side Slopes Combined</b>
<b>8 ft</b>	<b>No steeper than 2 H to 1 V</b>	<b>5 H to 1 V or Flatter</b>

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5/2011

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Revised Practice Standard Adoption Dates	Design Criteria Revised		
	Separation Distance in Feet From - -	Seasonal High Groundwater Table: Separation Distance From Pond Bottom	Liner Criteria: Soil type, Compaction, Permeability
1979, April PS- 425	Not specified	Do not construct to an elevation below the SHGWT unless considered a special design	Soils of slow to moderate permeability. Avoid gravel and shallow soils. If self-sealing is not probable, the storage pond shall be sealed by mechanical treatment or by the use of an impermeable membrane.
1987, February PS-425	300 ft from a neighboring residence, 200 ft from domestic well in an unconfined aquifer and 25 ft from water courses. All measured from outside toe of fill or top edge of pit pond	The operation and maintenance plan shall specify that the liquid level in the pond be maintained at least 6-in. above the ground water	No liner required for dense glacial till soils. GM, SM and ML materials may be used for a 12-in compact liner GC and SC materials may be used for a 9-in compacted liner CL and CH materials may be used for a 6-in compacted liner
1994, January PS-425	300 ft from any existing water wells unless aquifer evaluated for reduced distance	Do not construct below the SHGWT and shall have a properly designed and installed liner	1-ft minimum thickness, compacted soil liner of acceptable USCS soil material identified and listed as: CH, CL, MH, ML, and SM, SC, GM, GC if they contain more than 20% fines (passing #200 sieve)
2000, February PS-313	300 ft from any existing water wells unless aquifer evaluated for reduced distance	Pond bottom elevation shall be a minimum of 2 ft above SHGWT.  Depth to SHGWT shall be determined from soil features with the assistance of a soil scientist or from monitoring wells.	1-ft minimum thickness, compacted soil liner of acceptable USCS soil material identified and listed as: CH, CL, MH, ML, and SM, SC, GM, GC if they contain more than 20% fines (passing #200 sieve)
2001, June PS-313	300 ft from any existing water wells for storage pond unless aquifer evaluated for reduced distance.	Pond Bottom, Minimum 2 ft above SHGWT. SHGWT may be lowered by perimeter drains if feasible. Engineering Tech Note #7 (formerly agronomy Tech Note #42) shall be used to identify soil features for establishing the SHGWT.	Foundation permeability cannot exceed $1 \times 10^{-6}$ cm/s or it must be lined. All soil liners shall have a minimum compacted thickness of 1-ft. Compacted soil liner permeability must be equal to or less than $1 \times 10^{-6}$ cm/s.
2004, December PS-313	100 ft from any existing water wells. Aquifer evaluation required for variance but, must meet state and local regulations.	Pond Bottom, Minimum 2 ft above SHGWT. SHGWT may be lowered by perimeter drains if feasible and buoyant forces are considered.	1-ft minimum thickness of compacted soil liner. Permeability not to exceed $1 \times 10^{-6}$ cm/s

Message

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**From:** Winiecki, Eric [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=45FA974E305B49CD8AF513001A800543-WINIECKI, ERIC]  
**Sent:** 3/2/2013 12:10:17 AM  
**To:** Cox, Michael [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=cddd6a5bb3c2477183799ef56cdb464f-Cox, Michael]; Eaton, Thomas [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; MacDonald, Jennifer [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=507c2f57e60741b2b137cdf441c1c731-MacDonald Jennifer]; Fleming, Sheila [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=5d3e29dad0ec4cd98d87e1a142435c57-Fleming, Sheila]; Fuentes, Rene [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=3eb2a2e594934952b91ca7ade37262c5-Ruentes, Rene]; Jennings, Marie [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=ec4bd403e1994b80976542bb9458ae72-Jennings, Marie]  
**Subject:** Updated Lagoon Section  
**Attachments:** Lagoon-NRCSCriteriaRevisions.pdf; Yakima - Lagoon Responses (4).docx

Hi folks,

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Eric

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WASHINGTON NRCS - *DRAFT GUIDANCE*

5/2011

Page 27 of 29



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Message

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**From:** Cox, Michael [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=CDDD6A5BB3C2477183799EF56CDB464F-COX, MICHAEL]  
**Sent:** 3/2/2013 12:53:55 AM  
**To:** Winiecki, Eric [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=45fa974e305b49cd8af513001a800543-Winiecki, Eric]; Eaton, Thomas [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=10fa13022096442d83a97a2b7a699f06-Eaton, Thomas]; MacDonald, Jennifer [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=507c2f57e60741b2b137cdf441c1c731-MacDonald Jennifer]; Fleming, Sheila [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=5d3e29dad0ec4cd98d87e1a142435c57-Fleming, Sheila]; Fuentes, Rene [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=3eb2a2e594934952b91ca7ade37262c5-Ruentes, Rene]; Jennings, Marie [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=Ec4bd403e1994b80976542bb9458ae72-Jennings, Marie]  
**CC:** Cox, Michael [/O=EXCHANGELABS/OU=EXCHANGE ADMINISTRATIVE GROUP (FYDIBOHF23SPDLT)/CN=RECIPIENTS/CN=Cddd6a5bb3c2477183799ef56cdb464f-Cox, Michael]  
**Subject:** RE: Updated Lagoon Section  
**Attachments:** Yakima Comments Lagoon March 1 2013.docx

If you want to comment on the lagoon section please use this version. It is in the right format and will make easier to insert. I combined a few of the comments and responses but no change in the content.

Michael Cox  
Office of Environmental Assessment  
US EPA Region 10, 1200 Sixth Avenue, Suite 900  
Seattle, WA 98101  
206-553-1597  
cox.michael@epa.gov

---

**From:** Winiecki, Eric  
**Sent:** Friday, March 01, 2013 4:10 PM  
**To:** Cox, Michael; Eaton, Thomas; MacDonald, Jennifer; Fleming, Sheila; Fuentes, Rene; Jennings, Marie  
**Subject:** Updated Lagoon Section

Hi folks,

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Thanks,  
Eric

Message

---

**From:** Dennis McLerran [McLerran.DennisLNDU@usepa.onmicrosoft.com]  
**Sent:** 2/15/2013 12:40:15 AM  
**To:** Bob Sussman [Sussman.BobLNDU@usepa.onmicrosoft.com]; CN=James O'Hara/OU=DC/O=USEPA/C=US@EPA  
**CC:** Holsman, Marianne [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=332ee5ed102f4f65841285a4c06ea8b2-Holsman, Marianne]; Dunbar, Bill [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=133b66d6ab1b42108751e37b28176ec3-Dunbar, Bill (William) D.]; Thomas Eaton [Eaton.ThomasLNDU@usepa.onmicrosoft.com]; Opalski, Dan [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=8b5ed6410d934bf699a008a252791a55-Opalski, Dan]; Kowalski, Edward [/o=ExchangeLabs/ou=Exchange Administrative Group (FYDIBOHF23SPDLT)/cn=Recipients/cn=05111ec2f4314434a376f1c84c851820-Kowalski, Ed]; Joyce Kelly [Kelly.JoyceLNDU@usepa.onmicrosoft.com]  
**Subject:** Draft Letter Re: Yakima Valley to Washington NRCS  
**Attachments:** Yakima ltr to NRCS 2-14-13ver3.docx



Yakima ltr to  
NRCS 2-14-13ve...

Bob and Jim:

Here is a first cut at a letter from me to Washington NRCS Director Roylene Rides-at-the-Door.

Dennis